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OCTOBER 1958

LEAD-ALPHA AGE DETERMINATIONS OF GRANITIC ROCKS FROM ALASKA*

J. J. MATZKO, H. W. JAFFE, and C. L. WARING

ABSTRACT. Seventeen lead-alpha age determinations were made on zircon from eight granitic rocks of southeastern and central Alaska. The results of the age determinations indicate two periods of igneous intrusion, one about 100 million years ago during the middle of the Cretaceous period in southeastern and east-central Alaska, and another about 55 million years ago either very late in the Cretaceous or early in the Tertiary period in central Alaska. The individual ages determined on zircon from two rocks from southeastern Alaska and two from east-central Alaska gave results of 93, 103, 103, and 105 million years; those determined on four rocks from central Alaska gave results of 49, 53, 58, and 60 million years.

INTRODUCTION

Most of Alaska has been mapped on a reconnaissance basis only and definite age correlations are lacking in many areas. Many of the intrusive rocks that have been mapped in greater detail have been assigned indefinite ages because diagnostic fossils were not found in nearby sedimentary rocks, or because lithologic correlations could not be made with rocks of known age. As a means of obtaining the age or corroborating ages of the intrusive rocks that have been estimated from the geology, lead-alpha ages were determined on zircons concentrated from igneous rocks collected from several localities in Alaska (fig. 1).

Samples of granitic rocks from which the age determinations were made were collected by C. L. Sainsbury (written communication, 1957) from Tolstoi Point in southeastern Alaska, by Plafker (1956) from Turner Lake, near Taku Inlet in southeastern Alaska, by J. F. Seitz from Porcupine Creek in east-central Alaska, by White, Nelson, and Matzko (written communication, 1956) from Mount Fairplay in the Tanacross quadrangle, by White and Stevens (1953) from Birch Creek in the Ruby-Poorman district of central Alaska, by White and Killeen (1953) from Flat Creek and Chicken Creek in the Iditarod quadrangle, and by Matzko and Bates (written communication, 1957) from the Nixon Fork mines area, Medfra quadrangle. The age determinations were done by the Geological Survey on behalf of the Division of Raw Materials, U. S. Atomic Energy Commission.

These are the first age determinations based on radioactivity methods, made of minerals from Alaskan igneous rocks. This preliminary study indicates one period of igneous intrusion in southeastern and east-central Alaska about 100 million years ago during the middle of the Cretaceous period and another in central Alaska about 55 million years ago either very late in the Cretaceous or early in the Tertiary period. Ages determined on zircon from 7 of the 8

* Publication authorized by the Director, U. S. Geological Survey.

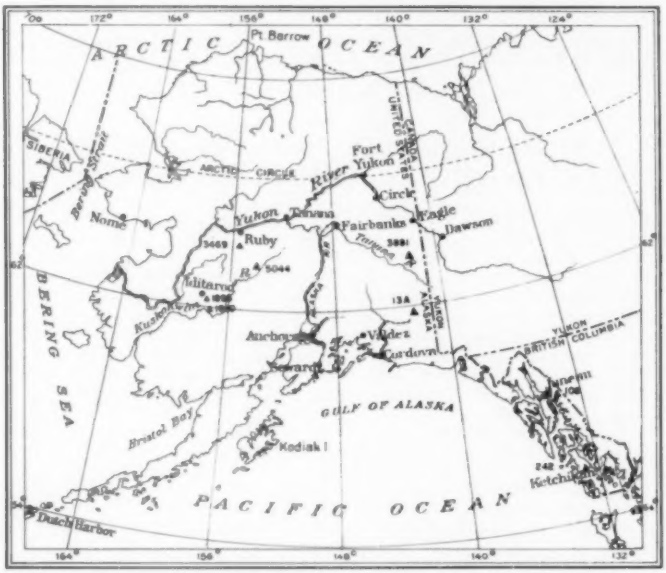

Fig. 1. Index map of Alaska showing sample locations.

rocks are in good agreement with age estimates made on the basis of geologic mapping; the age of the other is in disagreement.

Separation and purification of zircon for age determinations and alpha counts were made by J. J. Matzko and H. W. Jaffe. All lead determinations were made spectrographically by C. L. Waring. The method used in the determination of the age of the zircon has been described by Larsen, Keevil, and Harrison (1952), Lyons, Jaffe, Gottfried, and Waring (1957), and Quinn, Jaffe, Smith, and Waring (1957). The lead-alpha ages reported herein were calculated from the formula:

$$T = \frac{C \times Pb\ (ppm)}{\alpha/mg/hr}$$

where T = the age of the zircon in million years, Pb (ppm) = the total lead content in parts per million determined by emission spectrography, α/mg/hr = the total alpha activity due to the uranium + thorium, and C = 2485, a constant based upon an assumed Th/U ratio of 1.0. The estimated limit of error in the method is ± 10 percent or 10 million years, whichever is greater.

SOUTHEASTERN AND EAST-CENTRAL ALASKA

Tolstoi Point Geology.—The intrusive body at Tolstoi Point, Prince of Wales Island, Craig C-2 quadrangle, has been described by Warner, Goddard, and others (written communication) as a large stock-like intrusive. Porphyry dikes are cut by the stock and alkali andesite dikes intrude the stock. The

Tolstoi Mountain stock is composed mainly of light-gray, quartz-bearing diorite with medium-grained to coarse-grained granitoid texture. Wright (1915) has noted that the stock invades sedimentary strata of Devonian age. Buddington and Chapin (1929) suggest that the igneous rocks of Prince of Wales Island are satellitic intrusions of the Coast Range batholith which forms most of the mainland of southeastern Alaska. The age of the batholith is believed to be Late Jurassic or Early Cretaceous.

A calculation of the norm, according to the C.I.P.W. classification, and the chemical analysis from which the norm was derived is shown below for sample no. 242 from Tolstoi Point, collected by C. L. Sainsbury. In the classification, the rock falls into class 2, order 4, rang 3 and subrang 3. The chemical analysis was made by P. L. D. Elmore, K. E. White, and S. D. Botts, of the U. S. Geological Survey using the rapid rock analysis method.

No. 242	Weight percent		Norm percent		
SiO_2	52.8	Quartz	3.3		
Al_2O_3	15.9	Albite	26.2		
Fe_2O_3	2.4	Anorthite	26.4		
FeO	7.5	Orthoclase	6.1		
MgO	5.5	Hypersthene	18.4	Wollastonite	6.0
CaO	8.9	Diopside	11.9	Enstatite	13.7
Na_2O	3.1	Magnetite	3.5	Ferrosilite	10.6
K_2O	1.0	Ilmenite	1.8		
TiO_2	.98	Apatite	1.0		
P_2O_5	.44	Calcite	.2		
MnO	.21				
H_2O	1.4				
CO_2	.10				
Total	100.2		98.8		

A point-count modal analysis of the above rock shows the following:

	Percent
Plagioclase (andesine)	50.6
Quartz	6.8
Hornblende, green	32.4
Biotite-chlorite	7.4
Others	2.8
	100.0

"Others" include opaque minerals (magnetite and hematite), prehnite, zircon, sphene, apatite, clinozoisite, and pyroxene, in decreasing order of abundance.

The rock is a diorite with a hypidiomorphic granular texture and an average grain size of 1.5 mm. Its plagioclase is generally subhedral; tabular-like grains may be euhedral with a maximum length of about 2 mm; some

grains show abundant alteration to sericite and other grains appear relatively fresh. The alteration is believed to be primarily deuteric and not due to surface weathering. Quartz is predominantly interstitial and most quartz grains show undulatory extinction and secondary overgrowths. Schiller structure is shown on a few of the green hornblende grains. The hornblende is altered in part to biotite and to chlorite. Prehnite has developed in the biotite along cleavage lines which it tends to distort. Poikilitic quartz in minor amounts also is found in the biotite. Some of the biotite has been deformed as shown by warped cleavage traces, and drag effect on the biotite edges in contact with fresh plagioclase. Opaque mineral inclusions are abundant in the hornblende and biotite and some grains appear to replace sphene. A crystal of euhedral zircon in chlorite showed a strong pleochroic halo.

Age determination.—Age was determined on fresh, clear, doubly terminated zircon with no zoning and an index of refraction for omega of 1.92.

Sample no.	α/mg/hr	Pb (ppm)	Age (M.Y.)
242	142	5.8, 6.0	103

Turner Lake Geology.—In 1954, Plafker collected for age determination a sample from the Coast Range batholith at Turner Lake, near Taku Inlet, in the Taku River quadrangle of southeastern Alaska. An indefinite age of Late Jurassic or Early Cretaceous has been assigned to the Coast Range batholith in southeastern Alaska by Buddington and Chapin (1929, p. 253).

Plafker describes sample 106, from which the zircon was concentrated, as a medium-grained light-gray, hornblende-biotite granodiorite with a hypidiomorphic granular texture. His point-count modal analysis on the sample is shown below:

	Percent
Plagioclase (An 30-35)	52.3
Quartz	21.2
Biotite	15
Hornblende	5
Potassic feldspar	6.4
Others	.1
Total	100.0

"Others" are epidote and sphene that occur in trace amounts. The plagioclase is partly altered to sericite; biotite and hornblende are altered to chlorite. The average grain size of the granodiorite is 3.2 mm.

Age determination.—Age was determined on fresh, colorless, doubly-terminated, unzoned zircon with an omega index of refraction of 1.92.

Sample no.	α/mg/hr	Pb (ppm)	Age (M.Y.)
106	152	5.8, 5.6	93

Porcupine Creek Geology.—Porcupine Creek is in the Kuskulana River area of the Chitina Valley, east-central Alaska. The geology of the Chitina Valley was investigated by Moffit (1938), who distinguished two general groups of intrusive rocks in the area. These include an older group of predominantly gabbroic to dioritic rocks which intrude Permian and older rocks,

and a younger group of predominantly granitic intrusives which cut Mesozoic rocks. The granitic intrusives of the younger group are considered by Moffit to "range in age from earlier than late Upper Jurassic to post-Eocene". The largest bodies of granitic rock in the Chitina Valley are correlated with smaller satellitic intrusives that cut Cretaceous sediments. They are mapped as undifferentiated Cretaceous and Jurassic intrusives on the geologic map of Alaska compiled by Dutro and Payne (1957). A sample of one of the younger group of intrusive rocks, a granodiorite from Porcupine Creek in the McCarthy quadrangle was collected by James F. Seitz for a lead-alpha age determination. Moffit and Mertie (1923) described the granodiorites as granular, generally non-porphyritic rocks, composed essentially of quartz, plagioclase, orthoclase, hornblende, and biotite. Magnetite, apatite, and sphene are the accessory minerals. The ratio of plagioclase to orthoclase is 2:1.

Age determination.—A lead-alpha age was determined on fresh, euhedral, unzoned, zircon with an omega index of refraction of 1.92.

Sample no.	α/mg/hr	Pb (ppm)	Age (M.Y.)
13-A	283	11, 13	105

Mt. Fairplay Geology.—The large body of granitic rock at Mt. Fairplay in the Fortymile district, Tanacross quadrangle, and many smaller bodies nearby were mapped as Mesozoic in age by Mertie (1937, p. 215-216, and pl. 1). He acknowledges, however, that on the basis of stratigraphic evidence alone these granitic rocks can be mapped only as post-Paleozoic and pre-Tertiary. The sample collected for age determination from the western slope of Mt. Fairplay at milepost 29 on the Taylor Highway was a leucosyenite composed essentially of microperthite and subordinate albite-oligoclase. A point-count modal analysis of a thin section of the rock gave:

Perthite	59.2
Albite-oligoclase	35.8
Quartz	0.4
Others*	4.6
	100.0

* predominantly hornblende, biotite, sphene, opaque minerals, and zircon, in decreasing order of abundance.

The rock had an average grain diameter of 2 mm.

Age determination.—Three fractions of zircon, differing in magnetic response, were obtained by passing the concentrate through the Frantz magnetic separator at different amperage and tilt. The zircon concentrates consisted of whole and broken crystals with a squat habit, showing short prisms doubly terminated by the pyramids. Some of the crystals were almost equant in the absence of the more characteristic elongation of the prisms. The zircon crystals ranged in color from a pale buff in the least magnetic fraction to brown in the most magnetic fraction. Microscopic examination of the three fractions of zircon indicated that the brown color was due essentially to an unidentified brown material coating many of the zircon crystals to a varying degree, and to the more metamict state of some of the grains. Most of the zircon grains

were zoned, consisting essentially of alternating zones of fresh zircon with an omega index of refraction of 1.92 and optically continuous metamict zircon for which the omega index of refraction ranged from 1.86 to 1.90. Some of the grains also contained unidentified microscopic inclusions. The more magnetic fractions contained more of the metamict zones of zircon and showed a systematic increase in the lead content and alpha activity. The zircon was split into three fractions and then leached in 1-1 nitric acid for a period of 15 minutes; this treatment resulted in solution of the brown material coating most of the zircon grains with an accompanying loss of both lead and alpha activity. Splits of two of the samples were also leached in concentrated nitric acid with similar results. Lead-alpha age determinations made of the three fractions of zircon before and after nitric acid treatment gave the following results:

Sample no.	α/mg/hr	Pb (ppm)	Age (M.Y.)
3881-B (untreated)	1620	68	104
(1-1 nitric acid)	1134	45	99
3881-C (untreated)	1930	72	93
(1-1 nitric acid)	1476	63	106
(concentrated nitric acid)	1270	48, 52	98
3881-E (untreated)	2600	115	110
(1-1 nitric acid)	1594	72	112
(concentrated nitric acid)	1550	60, 63	99
Mean age			103
Standard deviation			6

Although a considerable amount of the alpha activity and lead was removed from each sample by the acid treatment, the ages remain in good agreement, suggesting a solution of thorite (isostructural with zircon) or metamict zones of zircon, rather than selective removal of uranium, thorium or lead. The average of the eight age determinations, 103 million years, corresponds to the Cretaceous period for which a range of 60-130 million years was indicated by the Committee on the Measurement of Geologic Time for 1949-50 (Marble, 1950).

CENTRAL ALASKA

Birch Creek Geology.—Mertie and Harrington (1924) describe three bodies of granite in the Ruby-Poorman district, west-central Alaska. One is on the east side of the Sulatna River, northeast of Poorman; another is at the head of Flint Creek; the third lies along the ridge east of Birch Creek, a tributary of Flint Creek. They describe the granite from the three areas as a yellowish-gray holocrystalline, coarsely granular rock, locally porphyritic, with phenocrysts of orthoclase or microcline, and mica, with a subordinate amount of oligoclase or oligoclase-albite.

Sample no. 3469, from which the zircon was concentrated, was collected by White and Stevens (1953), from disintegrated coarse-grained granite on Birch Creek, Ruby quadrangle. This granite has been mapped by Mertie and Harrington (1924) who designated it as Mesozoic principally on the basis of

freshness and similarity to some of the granites in the Yukon-Tanana region. No stratigraphic evidence is available, however, that would indicate the exact age of the granite, and the field evidence indicates only that it is post-Mississippian.

Age determination.—Age was determined on pale-brown zircon, essentially fresh, with an omega index of refraction of 1.92 and showing a slight amount of narrow optically continuous zoning.

Sample no.	α/mg/hr	Pb (ppm)	Age (M.Y.)
3469 (untreated)	1342	30	56
(concentrated nitric acid)	1342	29	54
(concentrated nitric acid)	1366	26, 27	48
Mean age			53

Flat Creek and Chicken Creek Geology.—White and Killeen (1953) noted that the quartz monzonite at the headwaters of Flat (sample no. 1895) and Chicken Creeks (sample no. 1810) in the Iditarod quadrangle, has two facies, light-colored and dark-colored, that do not appear to be gradational but show a distinct line of contact; the texture ranges from coarse to fine grained.

Mertie and Harrington (1924, p. 70) described the quartz monzonite in the Flat area as composed essentially of orthoclase, plagioclase, quartz, augite, biotite, and hornblende, with minor amounts of magnetite, apatite, and zircon. The plagioclase ranges from oligoclase to labradorite and is equal to the orthoclase in abundance. On the divide between the heads of Flat, Chicken, and Slate Creeks the orthoclase and plagioclase are graphically intergrown. White and Killeen (1953, p. 8) noted that the biotite from the quartz monzonite in this area shows numerous very pronounced pleochroic halos surrounding crystals of zircon and allanite(?).

The age of the quartz monzonite as determined from the field evidence is late Eocene or post-Eocene (Mertie and Harrington, 1924). The monzonite cuts sedimentary rocks, consisting of shale and sandstone of Late Cretaceous age; the monzonite is therefore post-Late Cretaceous.

Age determination.—Age determination was made on fresh, clear, pale-amber, doubly terminated zircon with index of refraction for omega of 1.92. No zoning was observed.

Sample no.	α/mg/hr	Pb (ppm)	Age (M.Y.)
1895	334	6.2, 7.0	49
1810	315	7.8, 7.4	60

Nixon Fork mines Geology.—Brown (1926, p. 124) describes the bedrock at Nixon Fork mines, Medfra quadrangle, as a monzonite that contains orthoclase and plagioclase feldspar, quartz, brown biotite, green hornblende, and minor amounts of apatite, iron oxides, and pyroxene. A porphyry, closely related in composition to the monzonite, reportedly occurs as a chilled border phase of the monzonite body and as dikes.

A point-count modal analysis is shown for sample no. 5044, on which the age was determined. The sample was collected by J. J. Matzko and G. D. Eberlein from the bedrock east of the Crystal Shaft mine.

	Percent
Perthite	53
Andesine	33
Hornblende, green	11
Others	2
Quartz	1
	100

"Others" include biotite (dark brown and showing strong absorption), apatite, opaque minerals, sphene, calcite, zircon, and hematite. Sericite in minor amounts accompanies the plagioclase, and kaolin is more common with the perthite. Hornblende has altered partly to biotite, and biotite has altered to chlorite. Some of the zircon embedded in the biotite shows pleochroic halos. Sphene commonly forms reaction rims around an opaque mineral, presumably ilmenite.

The average grain size is 1 mm, but plagioclase laths show a maximum size of 3 mm.

The age of the monzonite is believed by Brown (1926, p. 120) to be either Eocene or early Oligocene. In the area that was sampled, the monzonite cuts limestone of Paleozoic age, and sandstone, shale, and slate of Late Cretaceous age.

Age determination.—The zircons are similar to those from samples 1810 and 1895. They are fresh, clear, pale amber, doubly terminated, with no zoning detected, and have an index of refraction for omega of 1.92.

Sample no.	α/mg/hr	Pb (ppm)	Age (M.Y.)
5044	252	5.8, 6.0	58

SUMMARY

Six lead-alpha age determinations made on zircon from 4 intrusive rocks of central Alaska gave a mean age of 55 ± 5 million years (table 1). This is compatible with an early Tertiary or possibly a very late Cretaceous age, allowing for the limit of error of the lead-alpha method, which is estimated to be ± 10 percent or 10 million years, whichever is the greater. It should also be borne in mind that the division between the Cretaceous and Tertiary periods set at 60 million years by the Committee for the Measurement of Geologic Time (1949-50) and at 58 million years by Holmes (1947) is based upon concordant lead-uranium age determinations of 55 to 61 million years on pitchblendes from Colorado and may be subject to a small error. Evidence that the beginning of the Tertiary period may be younger than 58 to 60 million years in Canada is suggested by the work of Folinsbee and Ritchie (1957) on the Kneehills tuff member of the Edmonton formation, Alberta, Canada, which they have shown to be of Late Cretaceous (Lance) age. A coal seam above the tuff, overlain and underlain by Late Cretaceous dinosaur fossils, yielded a potassium-argon age of 52 million years on feldspar (R. E. Folinsbee, written

communication; and W. D. Ritchie, unpublished master's thesis, 1957). A lead-alpha age determined by Jaffe on zircon from the tuff immediately below the coal seam yielded a value of 53 million years. Intrusive rocks of middle Eocene age from the Bearpaw Mountains, Montana (Brown and Pecora, 1949) and from the Tintic and East Tintic districts, Utah (T. S. Lovering and H. T. Morris, written communication) yielded lead-alpha age determinations of 46 and 45 million years, respectively, as determined by Jaffe. From these data it is not possible to state whether the mean age of 55 million years obtained on the rocks of central Alaska is more consistent with a very late Cretaceous or very early Tertiary assignment. The results on three of the four rocks are nevertheless in good agreement with the geologic age of very late Cretaceous or early Tertiary assigned by Brown (1926) and Mertie and Harrington (1924) on the basis of field evidence. The age determined on the intrusive along Birch Creek, 53 million years, is not in as good agreement with the geologic age of Mesozoic estimated by Mertie and Harrington (1924). The Mesozoic age assignment, however, is questionable inasmuch as the stratigraphic evidence indicates only that the rock is post-Mississippian.

Eleven age determinations on zircon from intrusive rocks of east-central and southeastern Alaska gave a mean age of 102 $\pm$ 6 million years, compatible with a Cretaceous age assignment; the limits for the Cretaceous period are 60 to 130 million years, according to the Committee for the Measurement of Geologic Time (1949-50). Mertie (1937), Moffit (1938), Moffit and Mertie (1923), and Buddington and Chapin (1929), all considered these rocks to be Jurassic or Cretaceous in age, on the basis of field information. In all probability they are Cretaceous rather than Jurassic inasmuch as the mean age of 102 $\pm$ 6 million years is virtually the same as the mean age of 103 $\pm$ 6 million years determined on both zircon and monazite from four plutonic rocks of Baja California of early Late Cretaceous (post-Albian, pre-Maestrichtian) age (Silver, Stehli, and Allen, 1956). The lead-alpha age determinations of zircon from three of these rocks were 98, 102, and 112 million years; monazite from an additional intrusive gave an age of 99 million years. The writers share the view of Beveridge and Folinsbee (1956) and Larsen, Gottfried, Jaffe, and Waring (1954) that the principal North American Cordilleran intrusives were emplaced in Cretaceous rather than Jurassic time.

TABLE 1

Summary of lead-alpha age determinations of zircon from intrusive rocks of Alaska

Central Alaska

Rock	Sample no.	Locality	Age (million years)	
Granite	3469	Birch Creek, Ruby quadrangle	56 54 48	53
Quartz monzonite	1895	Flat Creek, Iditarod quadrangle	49	
Quartz monzonite	1810	Chicken Creek, Iditarod quadrangle	60	
Monzonite	5044	Nixon Fork Mines, Medfra quadrangle	58	

Mean age of central Alaskan rocks	55
Standard deviation	5
Tertiary period	1-60

Southeastern and east-central Alaska

Leucosyenite	3881	Mount Fairplay, Tanacross quadrangle	104	103
			99	
			93	
			106	
			98	
			110	
			112	
			99	
Granodiorite	106	Turner Lake, Taku River quadrangle	93	
Diorite	242	Tolstoi Point, Craig C-2 quadrangle	103	
Granodiorite	13-A	Porcupine Creek, McCarthy quadrangle	105	

Mean age of southeastern and east-central Alaskan rocks	102
Standard deviation	6
Cretaceous period	60-130

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THREE POLLEN DIAGRAMS FROM CENTRAL MASSACHUSETTS

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ABSTRACT. Three pollen diagrams are presented from central Massachusetts. The diagrams are believed to record the vegetational sequence from the time of retreat of pre-Valders ice to a time just previous to European colonization. They are interpreted to show that a narrow treeless belt along the edge of the retreating ice was rapidly colonized by forest. During Two Creeks time a cool-temperate, and perhaps dry, climate permitted the growth of a mixed spruce and deciduous forest. The climate during the Valders stadium became cooler and more moist, and brought about a change in the frequency of the forest species. An alternative interpretation, according to which a return of park-tundra vegetation, under conditions of intense solifluction, occurred in Valders time, is not favored by the weight of evidence. The succeeding pine phase has been divided into two zones, the second of which was probably warmer and drier than the first. The three deciduous forest zones described from other parts of New England have been demonstrated, although only one of the diagrams contains the entire sequence. The dominant pollen types in the three zones, starting with the earliest, are: oak and hemlock; oak, pine, and hickory; birch, oak, hemlock, and chestnut. The upper part of the pine zone and the first two deciduous forest zones are believed to represent the hypsithermal interval. On the basis of the bog plant pollen spectra, the beginning of the third deciduous forest zone is thought contemporaneous with the beginning of the Sub-Atlantic pollen zone in Europe.

INTRODUCTION

In recent years, attention has been directed to the vegetational history of glaciated regions of North America since the retreat of the ice. A knowledge of the earliest vegetation and its subsequent changes in species composition and frequency illuminates many biogeographical problems and increases our knowledge of the climate and soils of the past.

Analysis of the pollen and spores contained in organic deposits has proved an enormously useful technique in the study of vegetational history. Pollen diagrams from southern New England (Deevey, 1939, 1943) have demonstrated that, following the retreat of the ice, there was an interval when spruce was very abundant. The spruce was later replaced by pine, which in turn was replaced by three deciduous forest zones, characterized respectively by oak and hemlock, oak and hickory, and oak and chestnut. Recent studies in Connecticut have added more detail to this sequence. Leopold (1955, 1956b) has demonstrated that there was an interval of "steppe-like" vegetation immediately after deglaciation. At the southernmost site investigated, there is evidence that the forest advanced and then retreated again during this interval. The spruce phase earlier described by Deevey has been divided into four zones. Starting with the earliest, the zones are characterized by rising spruce percentages, a spruce maximum, a spruce minimum with higher percentages of pine and deciduous trees, and a second spruce maximum. The spruce minimum has been correlated with the warmest part of the Two Creeks interstadial, and the succeeding spruce maximum is thought to represent cooling associated with glacial advance during Valders time. This correlation is supported by radiocarbon dates. Apparently the climate in southern Connecticut during the Two Creeks interval was warm enough to permit the growth of mixed spruce, pine, and hardwood forests; during Valders time climatic cooling altered the species composition of the forest (Leopold, 1956b). The diagrams from Con-

necticut contrast sharply with the sequence in northern Maine (Deevey, 1951), Nova Scotia (Livingstone and Livingstone, 1958), and Michigan (Andersen, 1954), where there is evidence of more severe climate during Two Creeks and Valders time. It is thought that in northern Maine and Nova Scotia the park-tundra established during the Two Creeks interstadial was replaced by tundra during the cold interval associated with the Valders glacial advance. In Michigan there is evidence that the spruce forests of Two Creeks time were replaced by park-tundra.

The purpose of this investigation has been to study the vegetational history of central Massachusetts. On the basis of three pollen diagrams from the vicinity of the Harvard Forest it has been possible to outline the vegetational history of the area, to correlate it with sequences from other parts of New England, and to speculate on the factors which could have influenced the changes in vegetation that have occurred since the melting of the ice sheet.

ACKNOWLEDGMENTS

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PRESENT VEGETATION, CLIMATE, AND SOILS OF THE REGION

The three sites investigated are in the vicinity of the Harvard Forest, Petersham, Mass. (see fig. 1). This region now supports a complex, species-rich hardwood forest. Ecological studies (Bromley, 1935; Lutz and Cline, 1947; Stout, 1952) show that the forest is dominated by red oak (*Quercus rubra*), ash (*Fraxinus americana*), red maple (*Acer rubrum*), black birch (*Betula lenta*), and sugar maple (*A. saccharum*). White pine (*Pinus strobus*) and hemlock (*Tsuga canadensis*) occur frequently. On some north-facing slopes and in ravines sugar maple, yellow birch (*B. lutea*), beech (*Fagus grandifolia*), and hemlock are more common. On a few well-drained slopes with thin soils, the forest is dominated by white and black oak (*Q. alba* and *Q. velutina*), hickory (*Carya ovata* and *C. glabra*), and, formerly, chestnut (*Castanea dentata*).

The climate of the region (Rasche, 1958) is continental, despite the proximity (ca. 70 mi, 110 km) of the Atlantic Ocean. The prevailing winds are westerly, so that the ocean has little modifying effect. The mean January

Fig. 1. Map of southern New England. Arrow indicates location of sites investigated.

temperature is about 21°F (–6°C), and the mean July temperature 69°F (21°C), a mean annual range of almost 50°F (27°C). The climate is moist: the average yearly precipitation is about 40 in. (200 mm). The irregular topography results in a variety of microclimates. Studies indicate that local temperature variations within the Harvard Forest cover from one fourth to one half the variations among stations from southern to northernmost New England.

The bedrock underlying the Petersham region is acidic granite, gneiss, and schist. Its surface is irregular and the mantle of glacial debris is thin. In the valleys, and occasionally on slopes, there are gravelly outwash deposits. The uplands are mantled with sandy till; overlying this is a loam which may be in part of eolian origin. There is evidence that frost action has modified many of the slopes. The period of most active solifluction is thought to have occurred prior to the deposition of the loam (Raup, 1951; Stout, 1952). Although the age of the till is not known with certainty, it is probably of Cary or late Cary age (Flint, 1953; MacClintock, 1954; Denny, 1956a).

DESCRIPTION OF SITES

Tom Swamp.—Tom Swamp is a large sphagnum bog located in a north-south trending bedrock valley, apparently of pre-glacial origin (exact location 42°31′ N, 72°13′ W, elevation 750 ft.). The valley floor is lined with outwash, and there are gravel terraces along the slopes. Glacial meltwater from the north may have drained through the valley, depositing the outwash. It is possible that the valley was partially filled with stagnant ice during this period.

The bog is about 4 km long and 0.5 km wide. On the central portion of the bog black spruce, white pine, larch, highbush blueberry, and wild raisin

form a dense forest. To the north and to the south, the vegetation is more open. To the north, shrubs grade into a sedge meadow which is rapidly building into the open water of the artificially damned Riceville Pond. To the south, ericaceous shrubs and sphagnum are building a bog mat into the open water of Harvard Pond. The outlet here has also been dammed; early maps show the area as wet meadow (Raup and Carlson, 1941). It was not possible to survey the bog surface, because of the dense vegetation. However, there are small streams on the bog surface flowing north to the Riceville outlet, and flowing south to the Harvard Pond outlet. The presence of the two outlets and the drainage streams indicates that the bog may be slightly domed.

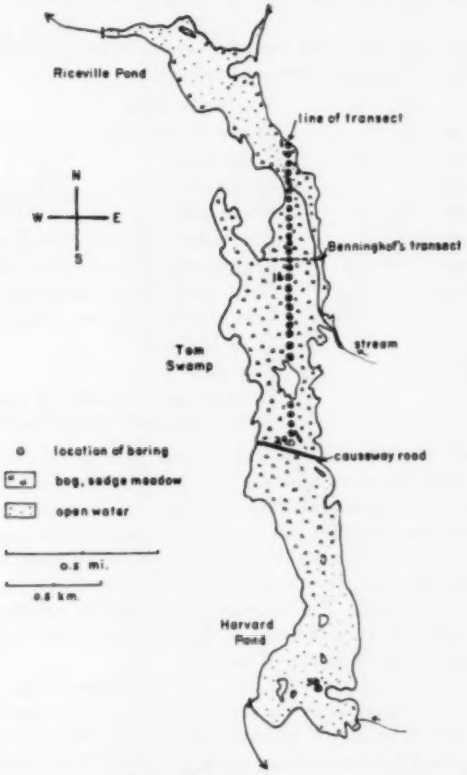


Fig. 2. Map of Tom Swamp.

A series of borings was made in an effort to determine the deepest part of the bog. A transect had previously been made across the bog by Prof. William S. Benninghoff (W. S. Benninghoff, personal communication). Accordingly, a trail was made due north and south from his deepest boring (see

fig. 2). Borings were made slightly less than 50 m apart. Samples were taken with a Hiller borer at 50 cm intervals, and peat identifications were made in the field. The resulting profile (plate 1) shows that forest occurs only on deeper parts of the bog. More hydrophytic vegetation occurs on the shallower parts of the basin. The Riceville sedge meadow is underlain by only a few meters of peat. Boring 30, made through the ice on Harvard Pond, showed that the peat here is also relatively shallow—only 1.5 m of water and 3 m of dy (lake mud) and peat were encountered. The classical hydrosere concept (cf. Weaver and Clements, 1938) is insufficient to explain this vegetational distribution. Apparently the ontogeny of Tom Swamp has been complex; possibly climatic changes have been a major factor influencing its development.

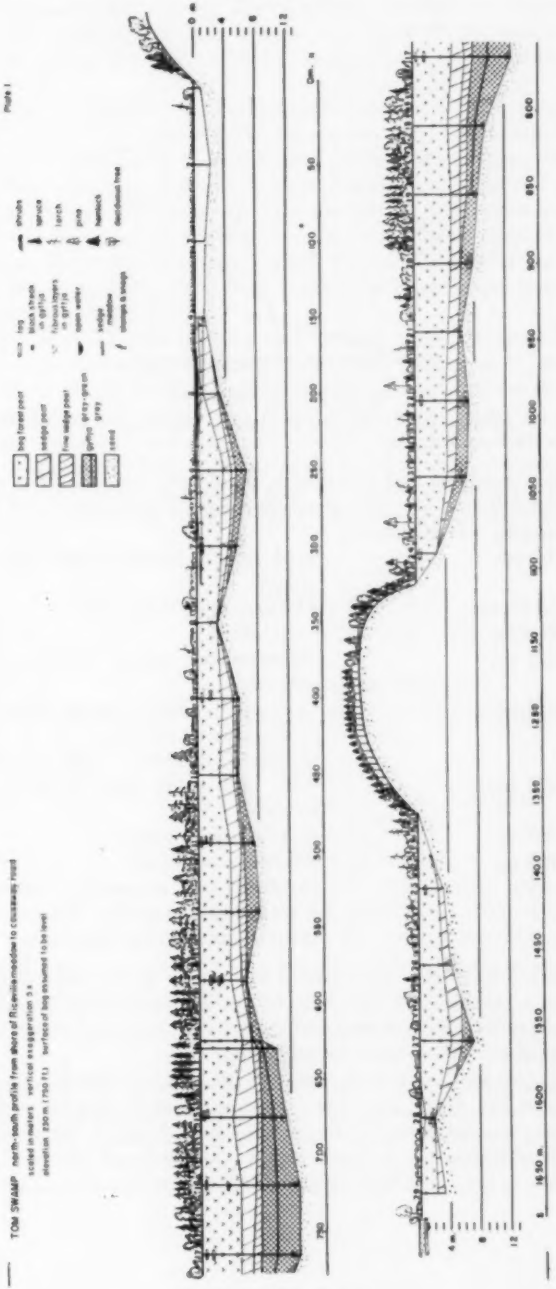
The samples used for pollen analysis were collected in August, 1955, from boring 16A. The stratigraphy here is as follows:

0-4.75 m	Bog forest peat
4.75-5.00 m	Coarse sedge peat
5.00-7.25 m	Sedge peat
7.25-7.55 m	Fine sedge peat
7.55-7.85 m	Sedge peat
7.85-8.05 m	Fine sedge peat; very wet from 7.98-8.05 m
8.05-9.90 m	Gray-green gyttja, slightly silty
9.90-12.50 m	Gray silty gyttja; scattered white specks from 10.85-11.55 m; black streaks from 11.70-12.50 m
12.50-12.91 m	Silty sand
12.91-13.00 m	Medium sand; it was not possible to penetrate deeper with the Hiller borer

The organic content of the sediments was determined by observing the percent dry weight lost upon ignition. Five of the eight samples tested were from half-meter bulk samples collected at boring 16C, located 1 m from boring 16A. Pollen from these samples was inspected to insure their contemporaneity with equivalent levels in boring 16A. The lower three samples tested were samples used for pollen analysis from boring 16A. The lowest sample weighed 3 g, the others 0.65 g each. The percent weight lost on ignition is shown in figure 3. The significance of the loss on ignition curve and the pollen zones indicated in figure 3 will be discussed below.

Pleasant St. Bog.—Pleasant St. Bog is located about 5 km (3 mi) NNW of Tom Swamp, in a small bedrock depression on a ridge near Athol, Mass. (exact location 42°34′ N, 72°14′ W, elevation 800 ft). The bog is ovoid, about 250 meters long, and has a maximum depth of about 5.5 m. In the summer of 1955 a road cut through the bog exposed the peat in open section. In most parts of the bog organic sediments directly overlie till, which, at least in the central portion of the bog, appears unweathered. It seems unlikely that an ice block would have persisted long in a depression of this size located near the summit of a ridge. Probably the sediments which have accumulated in the depression include the entire sequence since the retreat of the last ice sheet.

PLATE 1



Profile of Tom Swamp.

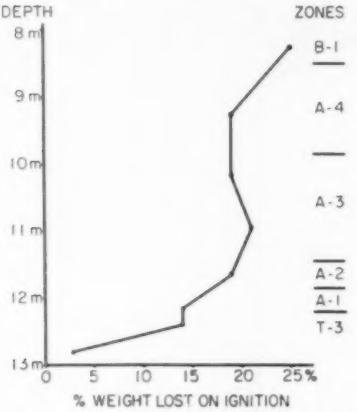


Fig. 3. Percentage loss on ignition of samples from Tom Swamp. Pollen zones are indicated to the right of figure.

Because slumping had occurred in the deeper parts of the bog, samples were collected from a peat face about 50 m east of the center of the bog. Here the stratigraphy was as follows:

0-119.5 cm	Dark brown, humified peat with logs and stumps
119.5-195 cm	Fibrous, unoxidized peat
195-218 cm	Gray gyttja
218-225 cm	Diatomaceous gyttja, slightly sandy and silty
225-240 cm	Gray gyttja, slightly sandy. A log, identified as *Populus*,[1] was found 5 cm to the east of the line of sampling at 230-240 cm depth
240-270 cm	Silty and sandy gray gyttja, with sand lenses
270-280 cm	Gray compact gyttja
280-290 cm	Blue-gray sandy silt
290 cm	Stratified sand, presumably outwash, of unknown depth, irregular surface, and maximum linear extent of about 6 m

Ignition loss studies were carried out on the lowest three samples. The lowest gyttja sample (280 cm) was 4.8 percent organic by weight; the silt and sand samples (285 cm and 290 cm), by contrast, lost only 0.3 percent and 0.4 percent of their weight upon ignition.

The poplar log found at the 230-240 cm level was dated by radiocarbon analysis. The log, 40 cm long and 10 cm in diameter, was divided into two parts, which were submitted to the U. S. Geological Survey Radiocarbon Laboratory, Washington, D. C., and to the University of Michigan Phoenix

[1] Identification by Prof. E. S. Barghoorn, Harvard University.

Project Radiocarbon Laboratory. They were dated by the acetylene method at 10,800 ± 250 years BP (W-361) (Crane and Griffin, 1958), and by the carbon dioxide method at 10,700 ± 800 years BP (M-413) (M. Rubin, personal communication).

Flint (1956) believes that the maximum advance of Valders ice in Wisconsin occurred 10,700 years ago. The log appears to be contemporaneous with the early part of this advance, or the latter part of the preceding interstadial (Two Creeks). The layer of gyttja in which it was found (zone A-3, discussed below) is probably of the same age. The position of the log was horizontal, and there was no evidence of disturbance of the overlying layer of diatomaceous gyttja. However, we cannot exclude the possibility that the log sank through still soft gyttja to its present position, and is younger than the sediments in which it was imbedded.

Gould's Bog.—Gould's Bog is a small hillside swale 2.5 km (1.5 mi) NE of Tom Swamp, and about 5.5 km (3.5 mi) SE of Pleasant St. Bog (exact location 42°32′ N, 72°11′ W, elevation 1110 ft). Most of the swale supports a red maple forest, but a small area of about 500 square meters is open bog, with sedges, sphagnum, and a few low shrubs. The sluggish drainage stream is artificially dammed by a low (1 m) earthen embankment. Probings showed the deepest point in the bog to be near the center of the open area. Here samples for analysis were taken in June, 1957. The stratigraphy was as follows:

0-27 cm	Wet fibrous peat
27-47 cm	Compact fibrous peat
47-82 cm	Brown amorphous peat with wood fragments
82-105 cm	Red-brown amorphous peat
105-122.5 cm	Compact fibrous peat with grass and sedge leaves
122.5-177 cm	Fine fibrous peat
177-247 cm	Brown gyttja, with dark gray streaks at 230 cm, and 234-239 cm
247-273 cm	Dark gray gyttja
273-300 cm	Black gyttja; the color is apparently due to a ferrous sulfide which oxidizes to a rusty red color upon exposure to air
300-318 cm	Black gyttja with gray silty streaks
318-325 cm	Gray silt with black streaks
325-350 cm	Light gray silty sand; it was not possible to penetrate deeper with the Hiller borer

Ignition loss data based on samples from this site are shown in figure 4. The curve shows a gradual increase in the organic content of the gyttja above the 300 cm level, culminating in an abrupt increase as the sediment changes to compact fibrous peat at the 125 cm level. The significance of these changes will be discussed below.

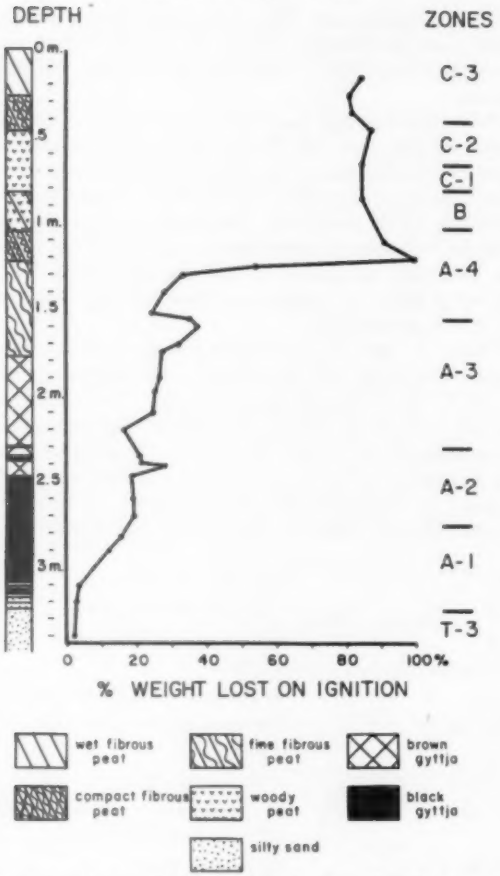


Fig. 4. Percentage loss on ignition of samples from Gould's Bog. Pollen zones are indicated to the right of figure.

TECHNIQUES

Samples were collected from Tom Swamp and Gould's Bog with a Hiller borer equipped with a 50 cm barrel. Two holes about 25 cm apart were used alternately. Samples were collected directly with a spatula from the open section of peat at Pleasant St. Bog. Preparation of samples followed in general the procedure suggested by Faegri and Iversen (1950). Samples were treated with 70 percent HF, 10 percent KOH, and acetolysis mixture. Basic fuchsin was used as a stain. A few of the fibrous peat samples from Gould's Bog were strained through a brass screen before acetolysis.

The sand collected from the lowest levels of Tom Swamp and Pleasant St. Bog contained very little pollen. Furthermore, the minerals reacted with HF

to form an insoluble residue. Warm 10 percent HCl followed by 50 percent H_2SO_4 was found effective in dissolving the residue. The bromoform flotation technique (Frey, 1951) was also used to separate the pollen from the residue. By these means it was possible to extract several hundred pollen grains from 2- or 3-g samples of sand. The sand beneath Gould's Bog was sufficiently rich in pollen that no special extraction techniques were necessary.

Samples were counted under a Zeiss apochromatic microscope. The oil immersion objective was used for all critical identifications, which were based on comparison with acetolyzed preparations of modern pollen. Percentages in samples from Tom Swamp and Pleasant St. Bog were calculated on the basis of a sum which includes all pollen except pollen of aquatic plants and pteridophyte spores. At Tom Swamp the percentage base includes at least 1000 grains, at Pleasant St. Bog, at least 700. At Gould's Bog, percentages were calculated on the basis of a sum which includes arboreal pollen (AP) only. At least 500 AP were counted in each sample.

DESCRIPTION OF THE POLLEN DIAGRAMS

The pollen diagrams from Tom Swamp, Pleasant St. Bog, and Gould's Bog are presented as plates 2, 3, and 4. Percentages of minor constituents not shown on the diagrams are given in tables 1, 2, and 3. For convenience in discussion the diagrams have been divided into pollen zones, which correspond in general to those used by Deevey (1939, 1943) and Leopold (1955). The characteristics of the zones, starting with the earliest, are outlined briefly here and discussed in more detail below.

T-3: Maximum of herbs, willow, alder, and *Juniperus-Thuja*.

A-1: Poplar and birch maximum. Herbs and shrubs decline, spruce rises.

A-2: Spruce maximum.

A-3: Spruce minimum. Deciduous tree maximum, followed by maximum of pine.

A-4: Spruce maximum.

B-1: Rising pine. Larch, birch, and alder maximum.

B-2: Pine maximum. Deciduous trees rising.

C-1: Oak and hemlock maximum, beech rising.

C-2: Hemlock minimum. Oak, pine, and hickory maximum.

C-3: Hemlock, birch, oak, and chestnut maximum, followed by oak, pine, and chestnut.

Zone T-3.—Only 30 to 40 percent of the pollen in this zone is arboreal. Of the trees, pine (*Pinus*) is most abundant, with spruce (*Picea*), birch (*Betula*), and oak (*Quercus*) of secondary importance. Curiously enough, at Tom Swamp and Pleasant St. Bog the percentages of arboreal pollen, particularly pine, are higher in the sand underlying the bog than they are in the organic gyttja. This is not the case at Gould's Bog, where the entire zone is represented by comparatively pollen-rich silty sand. At Gould's Bog the percentages of oak (7 percent of total pollen), ash (*Fraxinus*), elm (*Ulmus*), and hickory (*Carya*) are slightly higher than at the other sites.

TABLE 1

Tom Swamp 16A: Pollen Percentages not Shown on Diagram

depth in cm.	Armeria ?	Campanula	Chenopodiaceae	Coptis	Cruciferae	Impatiens	Liguliflorae	Polygonum scandens type	Rosaceae	cf. Dryas	Sanguisorba	cf. Saxifraga oppositifolia	Urticaceae	cf. Xanthium	Caprifoliaceae	Cornus	Rhus vernix	Viburnum	Castanea	Celtis	Juglans cinerea	Juglans nigra	Liquidambar	Equisetum	Liliaceae	Lycopodium selago	Lycopodium inundatum	Typha	Selaginella selaginoides	Potamogeton
625	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.16	.15	—	.10	—	—	—	—	.08	—	—	.15	—	—
645	—	—	.17	—	—	—	—	—	.17	—	—	—	—	—	—	—	—	—	—	—	.09	—	—	—	.09	—	—	.26	—	.09
665	—	—	—	—	—	—	.10	—	—	—	—	—	—	—	—	—	—	—	.10	—	—	—	—	—	—	—	—	—	—	—
685	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.08	—	—	—	.08	.08	—	—	—	—	—	.08	—	—
705	—	—	.09	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.09	.09	—	—	—	.19	—	—
725	—	—	.16	—	.08	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.09	.08	—	—	—	—	—	—	—	—	—
745	—	—	.09	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.09	—	—	.09	—	—	—	—	—	—	—	—	—
765	—	—	.10	—	—	.10	—	—	—	—	—	—	—	—	—	—	.20	—	—	—	—	—	—	—	—	—	—	—	—	.20
785	—	—	—	—	—	—	—	—	.09	—	—	—	—	—	—	—	—	.09	—	—	—	—	—	—	—	—	—	.28	—	.09
795	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.09	—	—	—	—	—	—	—	—	.09	—	.09
805	—	—	.07	—	—	—	—	—	.07	—	—	—	—	—	—	—	—	.07	—	—	.22	—	—	—	—	—	—	—	—	.07
815	—	—	.17	—	—	—	—	—	.17	—	—	—	—	—	—	.11	—	—	.17	—	—	—	—	—	—	—	.17	—	—
825	—	—	.11	—	—	—	—	—	.32	—	—	—	—	—	—	—	—	—	.11	—	—	—	—	—	—	—	.74	—	.42
835	—	—	—	—	—	—	—	—	.09	—	—	—	—	—	—	—	—	—	—	—	.09	—	—	—	—	.09	.18	—	.28
845	—	—	.08	—	—	—	—	—	.08	—	—	—	—	—	—	—	—	—	.26	—	.08	—	—	—	—	—	.42	—	—
855	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.09	—	—	—	—	—	—	—	.09	—	—	—	—	.09
865	—	—	—	—	—	—	—	—	.08	—	—	—	—	—	—	—	—	—	.08	—	—	—	—	—	—	—	—	—	.08
875	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.10	—	—	—	—	—	—	—	—	—

885	—	—	.08	.10	—	—	—	.08	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.08	—	—	—	.08	—	—
905	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.10	—	—	.10	—	.10
915	—	—	.10	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
925	—	—	—	—	—	—	.22	—	—	—	—	—	—	—	—	—	—	.11	—	—	—	—	—	.11	—	—	.07	—	.11
935	—	.07	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.14	—	.07	—	—	—	—
945	—	—	—	—	—	—	—	—	.09	—	—	—	.09	—	—	—	—	.09	—	—	—	—	—	—	.09	—	—	—	.09
955	—	—	—	—	—	—	—	—	.29	—	.10	—	—	—	.10	—	—	—	—	—	—	—	—	—	.10	—	—	—	—
975	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.09	—	.37	—	—
995	—	—	—	—	—	—	—	—	.37	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.09
1015	.06	—	.06	—	—	—	—	—	.18	—	—	—	—	—	—	—	—	—	—	—	.06	—	—	—	—	—	.30	—	—
1035	—	—	.28	—	—	—	—	—	.09	—	—	—	—	—	.09	—	—	—	—	—	—	—	—	—	—	—	.47	—	—
1055	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.28	—	.76
1065	—	—	.10	—	—	—	—	—	—	—	—	—	—	.10	—	—	—	—	.10	—	—	—	.10	—	—	—	.30	—	—
1075	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.10	—	—	—	—	.10	—	.10	—	—	—	.30	—	.30
1095	—	—	.10	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.10	—	—	—	—	—	—	—	.53	—	.10
1115	—	—	—	—	—	—	—	—	.23	—	—	—	—	—	—	—	—	—	—	—	.08	—	—	—	—	—	.38	—	.28
1135	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.54	—	—	—	.77	.15	.77
1145	—	—	.10	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.10	—	—	—	—	.29	—	—	—	.38	—	.58
1155	—	—	—	—	—	—	—	—	.09	—	—	—	—	—	—	—	—	—	—	—	—	—	.35	—	.18	—	—	—	.18
1165	—	—	.08	—	—	—	.10	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.08	.08	—	—	—	—	.08
1175	—	—	—	—	—	—	—	—	.09	—	—	.09	—	—	—	—	—	—	—	—	.09	—	.08	.09	—	—	.18	—	—
1185	—	—	—	—	—	—	—	—	.20	—	—	—	—	.10	—	—	—	—	—	—	—	—	—	.10	.10	—	.20	—	.10
1195	—	—	.09	—	—	—	—	—	.09	.09	—	—	—	—	—	—	—	.18	—	—	.09	—	.18	.09	.09	—	.27	—	.62
1215	—	—	.16	—	—	—	—	—	.16	—	.08	.08	—	—	—	—	—	—	—	—	—	—	.31	—	.08	—	.39	—	.70
1225	—	—	—	—	.18	—	—	—	.55	—	.09	—	—	—	—	—	—	.09	.20	—	—	—	.55	—	—	.09	.09	—	.82
1235	—	.10	—	—	—	—	—	—	.70	—	—	.20	—	—	—	—	—	—	—	—	—	—	.10	.20	—	—	.10	—	.50
1245	—	.10	.10	—	—	—	—	—	.93	—	—	—	—	—	—	—	—	—	—	—	.10	—	.31	—	—	—	—	—	.21
1255	—	—	—	—	—	—	—	—	.34	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.34
1265	—	—	—	—	—	—	—	—	.57	—	—	.57	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.28	—	1.14
1270	—	—	—	—	—	—	—	—	.72	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
1285	—	—	1.75	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—

Of the shrubs, alder (*Alnus*), willow (*Salix*), and the heaths (*Ericaceae*) are the most important. As willow tends to be underrepresented in pollen spectra (Iversen, 1947), this genus may have been the most abundant shrub. It is not known whether the Cupressineae pollen in zone T-3 represents an arctic shrub juniper (*Juniperus communis* var. *saxatilis*), a temperate shrub juniper (*J. communis* var. *depressa*), red cedar (*J. virginiana*), white cedar (*Chamaecyparis thyoides*), or arbor vitae (*Thuja occidentalis*). In any case, the percentages found here are low in comparison with the frequency of 10 percent reported from the late-glacial of Michigan (Andersen, 1954).

The dominant herbaceous types are sedges (Cyperaceae) and grasses (Gramineae). However, a wide variety of other flowering species are represented; the number of different pollen types found in each sample exceeds that found in samples from any of the other zones. The herbs found in zone T-3 fall into three categories: (1) genera or families which have both arctic and temperate species, (2) species which have a northern or arctic-alpine distribution, and (3) genera which occur in temperate regions only.

By far the largest proportion of the herbs identified fall in the first category. Many of the genera, such as *Artemisia*, *Thalictrum*, *Potentilla*, and *Rumex*, are characteristic of the Late-glacial of Europe.

Very few species can be put in the second category. *Saxifraga oppositifolia*, now arctic-alpine in distribution, has been tentatively identified from Tom Swamp and Pleasant St. Bog. A single grain of the *Polygonum bistorta* type was found at Gould's Bog. It most closely resembles *P. bistortoides*, which now grows in subalpine meadows in Newfoundland (Fernald, 1950). *Lycopodium selago* spores, distinguished tentatively on the basis of size from *L. lucidulum* (Leopold, 1955), were found in T-3 and the spruce zones. *L. selago* is arctic-alpine, but occurs on monutains as far south as North Carolina (Blomquist, 1934). Pollen of *Hippuris* cf. *vulgaris* was found at Gould's Bog in the spruce zones. This species now ranges in arctic regions and south to northern New England and northern New York state (Fernald, 1950). Microspores of *Selaginella selaginoides*, which has a similar range, were also found in the spruce zones. A few of the rosaceous pollen grains were tentatively identified as *Dryas*. Although *Dryas* is typically arctic-alpine, two of the American species range well into the boreal forest (Raup, 1947). *Sanguisorba canadensis*, although typically boreal, has a sporadic range far to the south (Fernald, 1950). On the whole, the evidence for an arctic element in the flora is rather tenuous. The species named above may indicate cool climates, but they cannot be used as conclusive evidence of a climate similar to that of the Canadian Arctic.

There is, however, good evidence for a temperate element in the herbaceous flora. *Polemonium*, for example, is a genus of temperate distribution in the northeast (except for a Greenland species). *Ambrosia*, *Plantago rugellii*, the Chenopodiaceae, and some of the species of the Tubuliflorae and Liguliflorae are now troublesome agricultural weeds in temperate regions. Their appearance in zone T-3 is analogous to the occurrence of a temperate element in the Late-glacial flora of Europe, where the temperate herbs disappeared almost

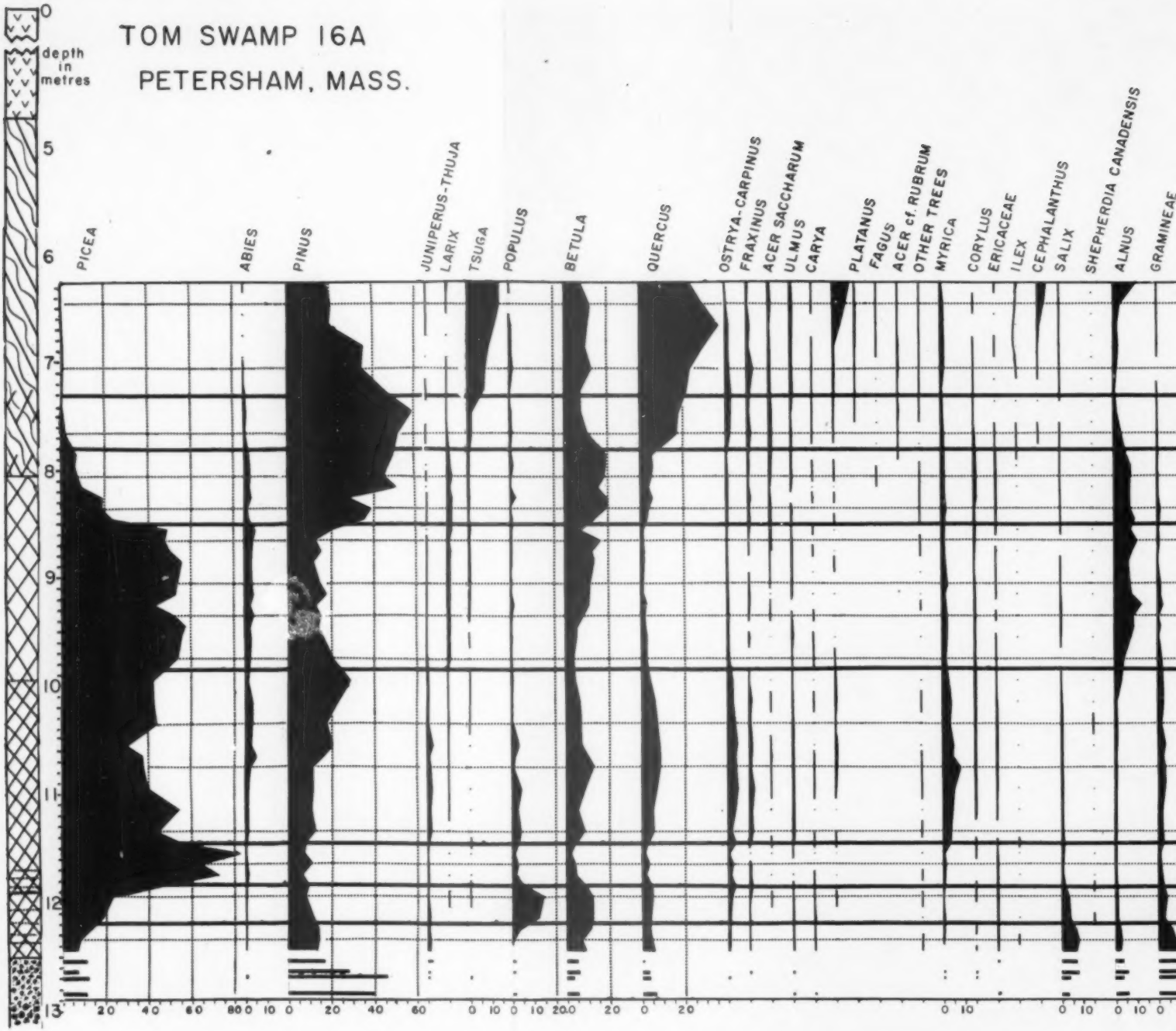
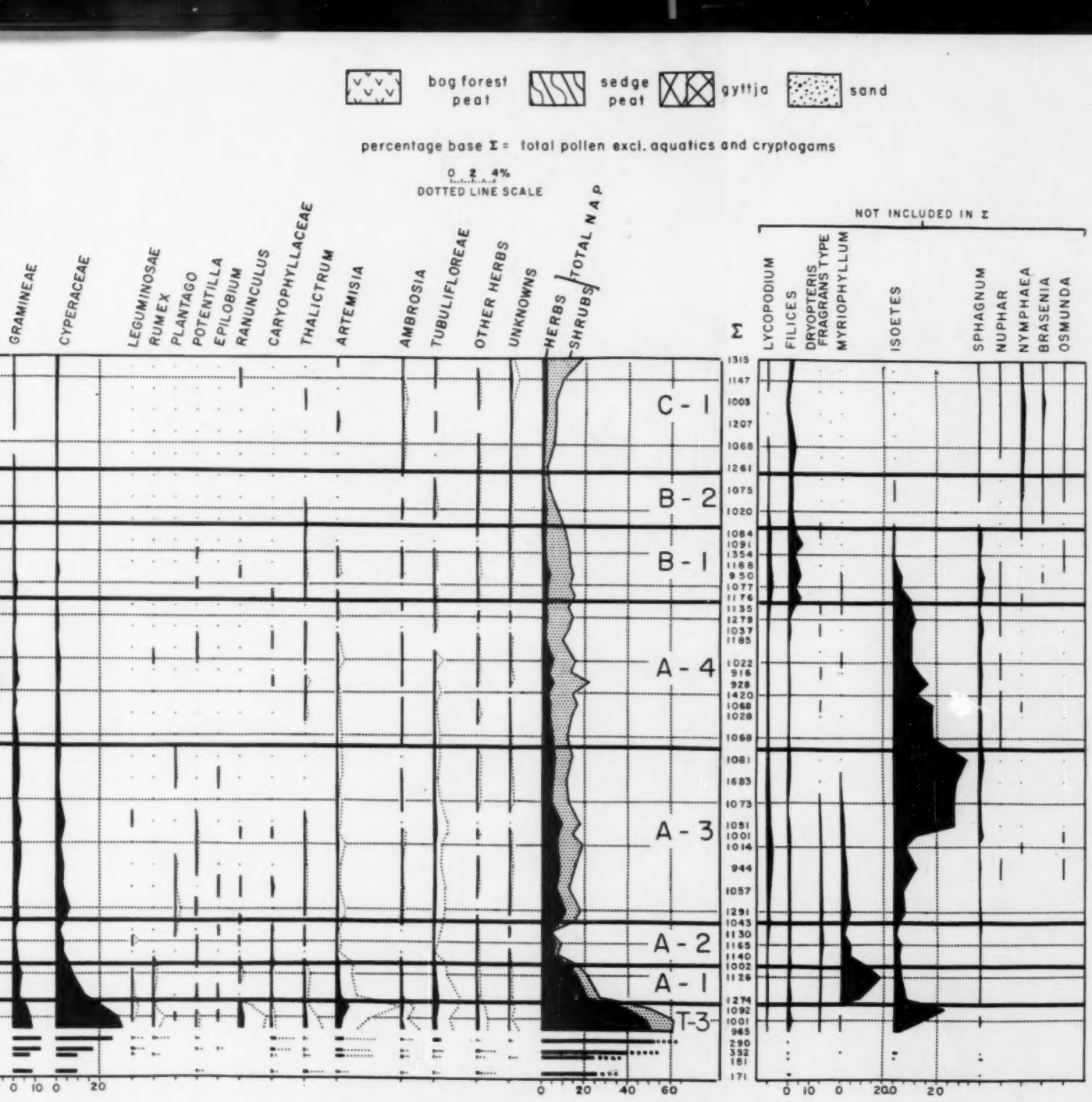


PLATE 2
Pollen diagram from Tom Swamp.


bog forest peat
sedge peat
gyttja
sand
percentage base Σ = total pollen excl. aquatics and cryptogams
0 2 4%
DOTTED LINE SCALE
GRAMINEAE
CYPERACEAE
LEGUMINOSAE
RUMEX
PLANTAGO
POTENTILLA
EPILOBIUM
RANUNCULUS
CARYOPHYLLACEAE
THALICTRUM
ARTEMISIA
AMBROSIA
TUBULIFLOREAE
OTHER HERBS
UNKNOWNS
HERBS
SHRUBS
TOTAL NAP
Σ
NOT INCLUDED IN Σ
LYCOPODIUM
FILICES
DRYOPTERIS FRAGRANS TYPE
MYRIOPHYLLUM
ISOETES
SPHAGNUM
NUPHAR
NYMPHAEA
BRASENIA
OSMUNDA
C - 1
B - 2
B - 1
A - 4
A - 3
A - 2
A - 1
T-3
1315
1147
1003
1207
1068
1261
1075
1020
1084
1091
1354
1188
950
1077
1176
1135
1279
1037
1185
1022
916
928
1420
1068
1028
1069
1081
1683
1073
1051
1001
1014
944
1057
1291
1043
1130
1165
1140
1002
1126
1274
1092
1001
965
290
352
181
171
0 10 0 20
0 20 40 60
0 10 0 200 20

entirely with the advent of Postglacial forests, to reappear much later as agricultural weeds (Iversen, 1954).

Zone T-3, then, is characterized by (1) high percentages of non-arboreal pollen; (2) a wide variety of herbaceous types, some of them of northern distribution; and (3) genera with high light requirements, such as *Epilobium*, *Campanula*, *Ambrosia*, the Chenopodiaceae, the Caryophyllaceae, *Thalictrum*, *Artemisia*, the Cupressineae (except arbor vitae), and many of the heaths and willows.

Zone A-1.—This zone is characterized by a maximum of poplar (*Populus*) and birch. Most of the poplar pollen is of the *P. balsamifera* type. Willow percentages remain high. The herbs are less frequent than in the preceding zone, but still abundant; the Tubuliflorae reach a maximum. The spruce curve begins to rise; pine percentages decrease. Pollen of *Sheperdia canadensis*, a shrub which is found in open spruce forest (Raup, 1947), occurs at all three sites.

Some interesting changes take place in the spectra of aquatic plants. At Tom Swamp, the *Isoetes* which characterized the preceding zone is replaced by *Myriophyllum*; at Pleasant St. Bog there is a low maximum of *Myriophyllum* and *Potamogeton*; at Gould's Bog the sediment changes from silty sand to gyttja, and *Isoetes* attains high percentages.

Zone A-2.—Spruce reaches a maximum of nearly 60 percent total pollen at Pleasant St. Bog, and 51 percent total pollen (64 percent AP) at Gould's Bog. At both sites oak, hop-hornbeam or ironwood (*Ostrya-Carpinus*), and ash percentages are rising. At Tom Swamp spruce makes up 83 percent of the total pollen; other pollen percentages have minima which appear to represent relative, rather than absolute, changes in frequency.

Zone A-3.—In this zone spruce declines to 30-40 percent. The pollen flora is rich. Poplar, juniper (cedar or arbor vitae?), *Dryopteris fragrans* type, and many of the herbs that characterized the preceding zones are still present. Club mosses (*Lycopodium*) are abundant. Oak, hop-hornbeam, ash, beech (*Fagus*), sugar maple (*Acer sacharrum*), elm, and a few other deciduous trees are at a maximum. Fir (*Abies*) percentages increase. Pine rises to a maximum of over 20 percent in the upper part of the zone.

The frequencies of a few of the pollen types vary from one site to another, and appear to reflect local, rather than regional changes. The birch maximum at Tom Swamp appears related to the *Myrica* maximum there. At Gould's Bog, the high frequency of sedges may represent a local marsh vegetation; the increase of grasses, Tubuliflorae, sphagnum and *Myrica* appears related to the change there from gyttja to fibrous peat.

Zone A-4.—In this zone spruce again reaches a maximum. Fir percentages remain constant; larch (*Larix*), birch, and alder percentages increase. Oak, hop-hornbeam, ash, elm, sugar maple, and beech frequencies decline to low levels.

At Tom Swamp the gyttja changes color from gray to gray-green. *Isoetes* and sphagnum are abundant. At Pleasant St. Bog, there is a layer of diatomaceous gyttja; *Isoetes* and sphagnum are at a maximum; pollen preservation is poor. At Gould's Bog, *Myrica*, alder, grasses, Tubuliflorae, *Potentilla*, sphag-

TABLE 2

Pleasant St. Bog: Pollen Percentages not Shown on Diagram

depth in cm.	Astragalus alpinum	Batrachium	Chenopodiaceae	Epilobium	cf. Gentiana	Liguliflorae	cf. Oxyria digyna	Rosaceae	cf. Dryas	Sanguisorba	cf. Saxifraga	Urticaceae	Xanthium	Cornus	Acer cf. rubrum	Carya	Castanea	Celtis	Juglans cinerea	Nyssa	Platanus	Tilia	Ulmus	Equisetum	Eriocaulon	Lycopodium inundatum	Lycopodium selago	Osmunda	Selaginella selaginoides
110	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.60	—	—	—	—	—	.12	—	.36	—	—	—	—	—	—
115	—	—	—	—	—	—	—	.21	—	—	—	—	—	—	.21	.21	—	—	.10	.10	—	.10	1.05	—	—	.21	—	—	—
120	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.28	.41	—	.28	.14	—	.28	.14	.83	—	—	.41	—	—	—
130	—	—	—	—	—	—	—	.12	—	—	—	—	—	—	.25	—	.12	—	—	—	.12	.12	1.01	—	.12	—	—	—	—
140	—	—	—	—	—	—	—	.34	—	—	—	—	—	—	.45	.34	.11	.11	—	—	—	—	.67	—	.11	—	—	—	—
150	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.64	.13	.26	—	—	—	.13	—	1.03	—	—	—	—	.13	—
155	—	—	.27	—	—	—	—	.18	—	—	—	—	.09	—	.18	.09	—	—	—	—	—	—	.55	—	—	—	—	—	—
160	—	—	—	—	—	—	—	.12	—	—	—	—	—	—	—	.12	—	—	—	—	.25	—	2.12	—	—	.25	—	—	—
166	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.59	—	.24	—	—	—	—	—	.59	—	—	—	—	—	—
171	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.27	—	—	—	—	—	.18	—	1.52	—	—	—	—	—	—
177	—	—	—	—	—	—	—	—	—	—	—	—	—	.06	.34	.06	.06	—	—	—	.11	—	.96	—	—	—	—	—	—

182	.10	—	—	—	—	—	—	.10	—	—	—	—	—	—	.39	.20	—	—	—	—	.10	—	.98	—	—	—	—	—	—
187	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.09	.09	—	—	.18	—	—	—	.72	—	—	—	—	—	—
195	—	—	.08	—	—	—	—	—	—	—	—	—	—	—	.33	.04	.04	—	.08	—	—	—	.70	—	—	—	—	.04	—
200	—	.16	—	—	—	—	—	.08	—	—	—	.08	—	—	.17	—	.08	—	.17	—	—	—	.60	—	—	—	—	—	—
210	—	—	—	—	—	—	—	.20	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.27	—	—	—	—	—	—
215	—	—	—	—	—	—	—	.47	—	—	—	—	—	—	—	.16	—	—	—	—	—	—	.62	—	—	—	—	—	—
220	—	—	—	—	—	—	—	.14	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.14	.14
225	—	—	.16	—	—	—	—	.31	—	—	—	—	—	—	—	.08	—	—	—	—	—	—	.55	—	—	—	—	—	—
230	—	—	—	—	—	—	—	.45	—	—	.11	—	—	—	—	—	—	—	—	—	—	—	.23	.11	—	—	—	—	—
235	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.10	—	—	—	—	—	—	.40	—	—	—	—	—	—
245	—	—	—	—	.15	—	—	—	—	—	—	—	—	—	—	.15	—	—	—	—	—	—	—	—	—	—	—	.15	—
250	—	—	—	.29	—	—	.14	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.14	—	—	—	.14	—	—
255	—	—	.13	—	—	—	—	.13	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
260	—	—	—	—	—	.12	—	.12	—	—	—	—	—	—	—	.12	—	—	—	—	—	—	—	—	—	—	.12	—	—
265	—	—	.11	—	—	—	—	.11	—	—	—	—	.11	—	—	—	.11	—	—	—	—	—	—	.11	—	—	—	—	—
270	—	—	—	—	—	—	—	1.69	—	—	—	—	.15	—	—	.15	—	—	—	—	—	—	—	—	—	—	—	—	—
275	—	—	—	—	—	—	—	.29	.29	.29	—	—	—	—	—	—	—	—	—	—	—	—	.15	—	—	—	—	—	—
280	—	—	—	—	—	—	—	1.10	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
285	—	—	—	—	—	—	.15	—	.15	—	.15	—	.15	—	—	—	—	—	.15	—	—	—	.30	—	—	—	—	—	—
290	—	—	—	—	—	—	—	.29	—	—	—	—	—	—	—	.58	—	—	—	—	—	—	.29	—	—	—	—	—	—

num, and ferns (Polypodiaceae) reach a maximum; *Isoetes* declines in frequency. As the peat becomes compact and fibrous, there is a strong maximum of sedge pollen.

Zones B-1 and B-2.—Zone B is characterized by a maximum of pine. At Tom Swamp and Pleasant St. Bog, where there are more samples from this zone, the pine maximum has been subdivided into two zones. In zone B-1, spruce and fir decline rapidly, pine rises, and oak and other deciduous trees begin to increase in frequency. There is a maximum of birch, alder, and larch. Club mosses, ferns, and sphagnum are abundant. At all three sites there is a change from more aquatic to a more terrestrial type of peat. At Gould's Bog, aquatic plant pollen is entirely absent; apparently the alder, ferns, and sphagnum are part of the bog vegetation.

In zone B-2, larch, birch, and alder percentages decrease, and oak and other deciduous trees become more abundant. Hemlock (*Tsuga*) frequencies begin to rise. At Gould's Bog there is a maximum of ferns (*Polypodiaceae* and *Osmunda*), and of the bog shrubs buttonbush (*Cephalanthus*), sumac (*Rhus*), and black alder (*Ilex*).

Zone C-1.—Zone C-1 is the first zone that is dominated by pollen of deciduous trees. Hemlock and oak are at a maximum. Sugar maple, ash, elm, and sycamore (*Platanus*) are present. Basswood (*Tilia*), hackberry (*Celtis*), and black walnut (*Juglans nigra*) occur in low frequencies (these three genera also occur in the spruce zone at Gould's Bog). Beech rises to a maximum along the C-1—C-2 border.

At Pleasant St. Bog there is a change from fibrous peat to bog forest peat in the upper part of this zone; the rising alder, birch, and Tubuliflorae percentages appear to reflect this change. At Tom Swamp, as well, the frequencies of bog plants such as buttonbush, sumac, and alder indicate that the bog was filling in rapidly. Apparently the maximum of birch and black gum (*Nyssa*) at Gould's Bog reflects a similar change.

Zone C-2.—Samples from this zone and zone C-3 were analyzed at Gould's Bog only. Consequently it is not known to what extent the pollen frequencies record local rather than regional vegetational changes. Zone C-2 is characterized by a hemlock minimum. Sugar maple, elm, and possibly basswood are also at a minimum. Hickory, which is so characteristic of this zone further south (Deevey, 1939, 1943, 1948), reaches a maximum frequency of 2 percent AP. Oak, followed by pine, has a strong maximum.

Zone C-3.—Zone C-3 is characterized by a return of hemlock, sugar maple, and elm, and a sudden rise in chestnut (*Castanea*). Ash also increases in frequency. Birch reaches a maximum in the lower part of the zone, but in the upper levels birch and hemlock decline and pine rises to a maximum. The sediment at Gould's Bog changes from bog forest peat to fibrous peat. Above the change, pollen of local bog plants (including spruce?) becomes abundant. No agricultural indicators, such as maize pollen or the pollen of European weed species, were found. Consequently the uppermost sample is thought to date from late precolonial time.

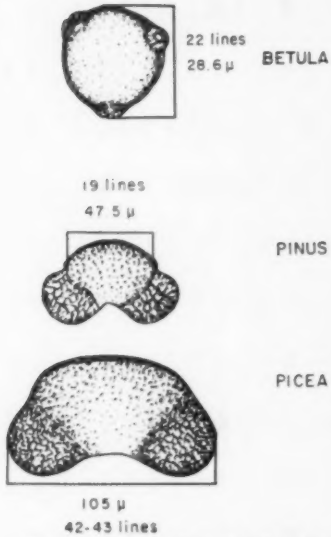


Fig. 5. Scale drawings of pollen from the 8.25 m. level of Tom Swamp, boring 16A. The black line indicates the dimension of the grain measured.

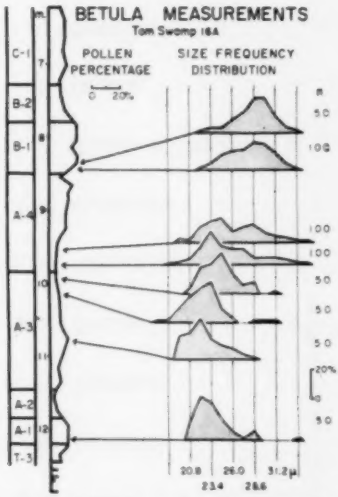


Fig. 6. Size-frequency distributions of birch pollen in 8 samples from Tom Swamp, boring 16A.

MEASUREMENTS OF POLLEN SIZE

In an attempt to make species identifications, pollen of birch, pine, and spruce was measured in samples from Tom Swamp. The method in each case was to use thick glycerine mounts which facilitated rotation of the grains. The delta (distance between ocular lines) used for birch measurements was 1.3 μ, for pine 2.5 μ, for spruce 5μ. The way in which the micrometer was positioned over the grains is shown in figure 5. Only samples in which preservation was excellent were used. Pollen which was broken, distorted, or poorly preserved was not measured.

Birch.—It is clear from the size frequency data (see fig. 6) that at least two types of birch are present. A type with small to medium-sized pollen (mode at about 23 μ) predominates in zones A-1 and A-3, and is replaced gradually in zone A-4 by a type with large pollen (mode at about 29 μ). The presence of bimodal curves in zone A-4, when both types are present, indicates that these size changes are not due to differential changes incurred during fossilization. A less complete series of measurements in four samples from Pleasant St. Bog confirm these results. We can only speculate as to which species are represented. The available data on the pollen size of the northeastern American birches (see Leopold, 1956a) shows that the size distributions of many species overlap, and that the size of different individuals of the same species may vary considerably. The shrub birches and at least one of the tree birches have small to medium-sized pollen, and the tree birches have medium-sized to large pol-

TABLE 3

Gould's Bog: Pollen Percentages not Shown on Diagram

depth in cm.	Celtis	Juglans cinerea	Juglans nigra	Cornus	Corylus	Viburnum	Vitaceae	Chenopodiaceae	Cruciferae	Galium	Labiatae?	Liguliflorae	Plantago	Polygalaceae?	Umbelliferae	Hippuris cf. vulgaris	Liliaceae	Nymphaea	Potamogeton
5		.35			.18	.18		.18		.18	.18								
15		.20	.10			.10				.20			.30						
20		.36			.12				.24		.12	.09							
25					.31				.31			.09	.31						
30					.18	.18		.19		.19			.19				.31		.38
35		.19			.19	.57		.13											
40			.26		.13														
45		.08										.09							.09
55					.09														
60									.07										
65								.31											
70		.16																	
75			.09		.09	.09		.09						.09					
80		.15																	
85					.16				.16										
95					.76			.13			.13								
102																			
105																			

110	—	—	—	—	.22	—	—	—	—	—	—	—	—	—	—	—	—	—	—
115	—	—	—	—	—	—	—	—	—	.35	—	—	—	—	—	—	.17	—	—
120	—	—	—	—	.18	—	—	—	—	.18	—	—	—	—	—	—	—	—	—
125	—	—	—	—	.41	.41	—	—	—	—	—	—	—	—	—	—	—	—	—
135	—	—	—	.10	.20	—	—	—	—	—	—	—	—	—	—	.20	—	.20	—
145	—	—	—	—	.29	—	—	—	—	.14	—	—	—	—	—	—	—	—	—
155	—	—	—	.14	.27	—	—	—	—	—	—	—	—	—	—	—	—	—	—
165	—	—	—	—	.34	—	—	—	—	—	—	—	—	—	—	—	—	—	—
170	—	—	—	—	—	—	—	.14	—	—	—	.29	—	—	—	—	—	—	—
180	—	.11	—	.34	.34	—	—	—	—	—	—	—	—	—	—	—	—	—	.11
185	—	—	—	—	—	—	.12	—	—	—	—	—	—	—	—	.12	—	—	—
195	—	—	—	—	.15	.15	—	—	—	—	—	—	—	—	.15	—	—	—	—
205	—	—	—	.16	.49	—	—	—	—	—	—	—	—	—	—	—	—	—	—
215	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
225	—	—	—	—	—	—	—	—	—	.23	—	—	—	—	—	—	—	—	—
235	—	—	—	—	.19	—	—	.19	—	—	—	—	—	—	—	—	—	—	.19
239	—	.16	—	—	.31	—	—	—	—	—	—	—	—	—	.31	.16	—	—	—
245	—	—	—	—	.17	—	—	—	—	—	—	—	—	—	.17	—	—	—	—
252	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	.16	—	—	—
255	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
265	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
270	—	—	—	—	.31	.31	—	—	—	—	—	—	—	—	—	—	—	—	.13
275	.18	—	—	—	.18	—	—	.18	—	—	—	—	—	—	—	—	—	—	—
285	—	—	—	—	.14	—	.14	—	—	.14	—	—	—	—	—	—	—	—	—
295	—	—	—	—	.16	—	—	—	—	—	—	—	—	—	—	.22	—	—	.16
298	—	—	.15	—	—	—	—	—	—	.15	—	—	—	—	—	—	—	—	—
305	—	.18	—	—	—	—	—	.18	—	—	—	—	—	—	.18	—	—	—	—
315	.18	—	—	—	—	.18	—	—	—	—	—	—	—	—	—	—	—	—	—
325	—	—	—	—	—	—	—	—	—	—	—	—	.27	—	—	.54	—	—	—
335	—	.33	—	—	—	—	—	—	—	—	—	—	—	—	.33	—	—	—	—
345	—	—	—	—	1.37	—	—	.34	—	—	—	—	—	—	—	—	—	—	—

len (Leopold, 1956a). If the absolute size of modern and fossil pollen can be compared, the 29 μ mode may represent tree birches with large pollen, such as yellow birch (*Betula lutea*) and white birch (*B. papyrifera*). The 23 μ mode may represent a shrub birch with small pollen, such as *B. glandulosa*, or gray birch (*B. populifolia*), a tree birch with small pollen. The data indicate, therefore, that tree birches are present in A-4 and predominate in B-1, and that shrub birch or gray birch may be present in zones A-1, A-3, and A-4.

Pine.—There are also two modes in the size frequency distributions of pine pollen (see fig. 7). In A-3, A-4, and B-1 the population is dominated by a pine with a pollen size of about 47 μ. In A-4 a small proportion of the population has pollen with a mode at about 60 μ. The latter type increases rapidly in late B-1 and predominates in zone B-2. The slight shift in the mode of the small-sized pollen to a larger size-class in early A-4 is thought to be due to a difference in preservation; the birch at these levels also has a larger absolute size (cf. Wenner, 1953).

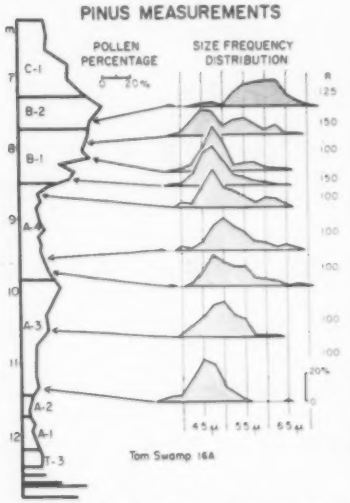


Fig. 7. Size-frequency distributions of pine pollen in 9 samples from Tom Swamp, boring 16A.

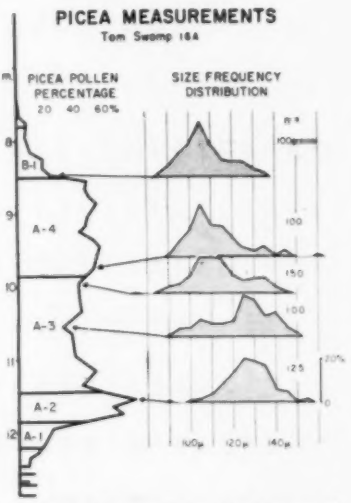


Fig. 8. Size-frequency distributions of spruce pollen in 5 samples from Tom Swamp, boring 16A.

Measurements of modern pollen indicate that certain of the eastern species of pine may be distinguished on the basis of pollen size (Cain and Cain, 1948; S. A. Cain, personal communication). These authors have found that the mean size of the internal diameter of acetolized pine pollen is as follows:

Pinus rigida (10 collections) ..... 55.16 μ, σ 3.12 μ
P. strobus (9 collections) ..... 50.38 μ, σ 4.03 μ
P. resinosa (10 collections) ..... 46.00 μ, σ 2.85 μ
P. banksiana (12 collections) ..... 45.06 μ, σ 2.83 μ

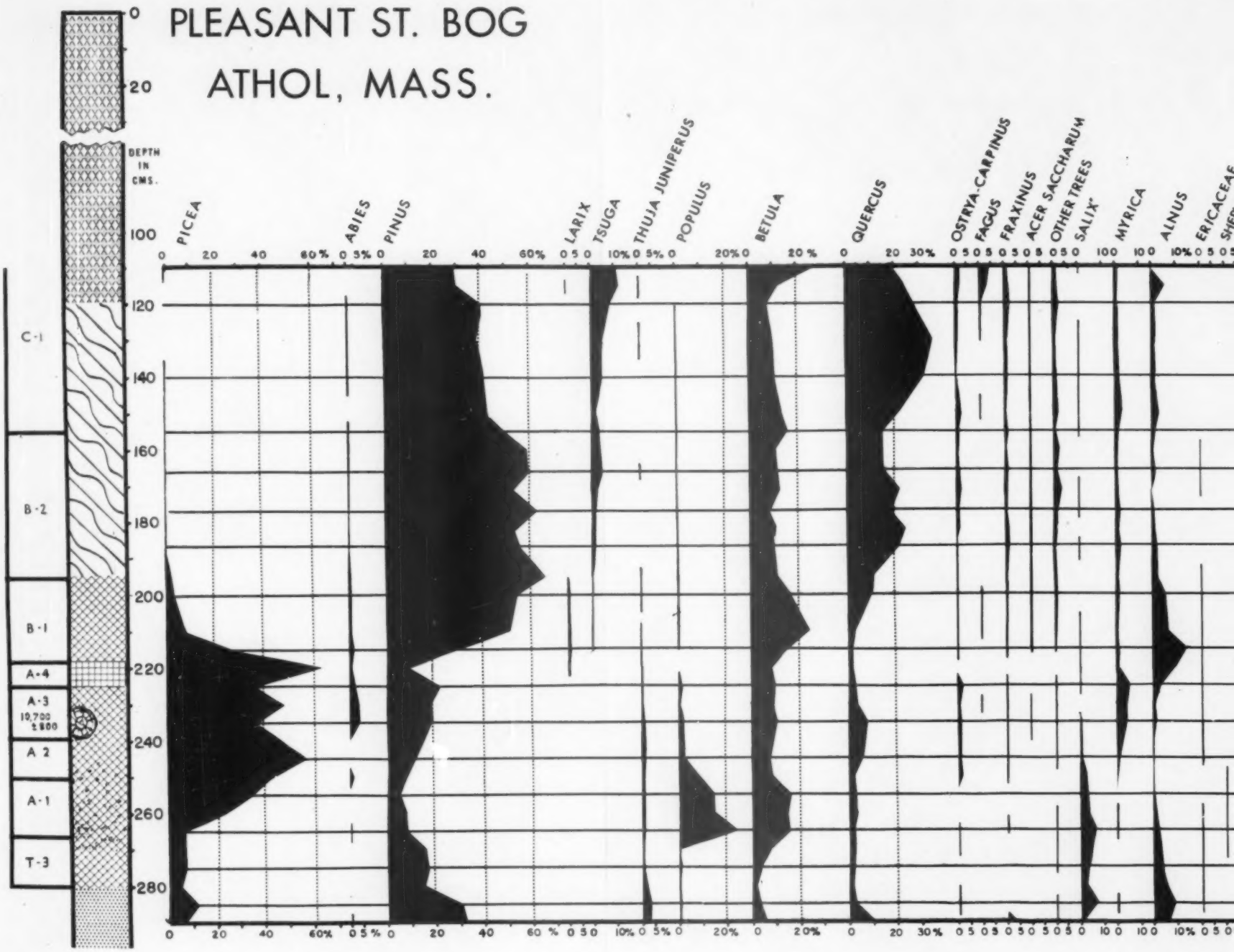
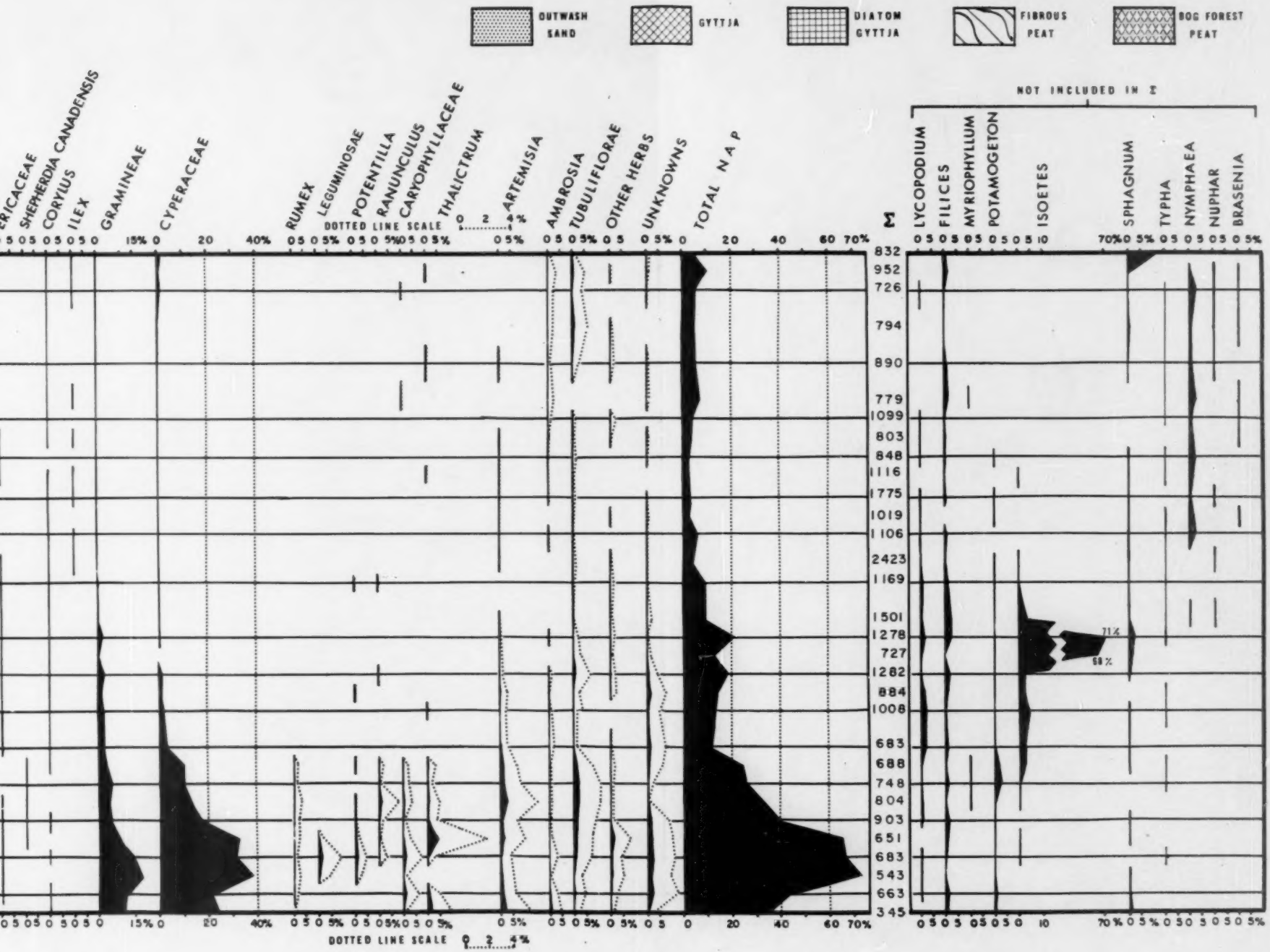


PLATE 3
Pollen diagram from Pleasant St. Bog.


OUTWASH SAND
GYTTJA
DIATOM GYTTJA
FIBROUS PEAT
BOG FOREST PEAT
ERICACEAE
SHEPHERDIA CANADENSIS
CORYLUS
ILEX
GRAMINEAE
CYPERACEAE
RUMEX
LEGUMINOSAE
POTENTILLA
RANUNCULUS
CARYOPHYLLACEAE
THALICTRUM
ARTEMISIA
AMBROSIA
TUBULIFLORAE
OTHER HERBS
UNKNOWNS
TOTAL NAP
DOTTED LINE SCALE
NOT INCLUDED IN Σ
LYCOPODIUM
FILICES
MYRIOPHYLLUM
POTAMOGETON
ISOETES
SPHAGNUM
TYPHA
NYMPHAEA
NUPHAR
BRASENIA
Σ
832
952
726
794
890
779
1099
803
848
1116
1775
1019
1106
2423
1169
1501
1278
727
1282
884
1008
683
688
748
804
903
651
683
543
663
345
71%
58%

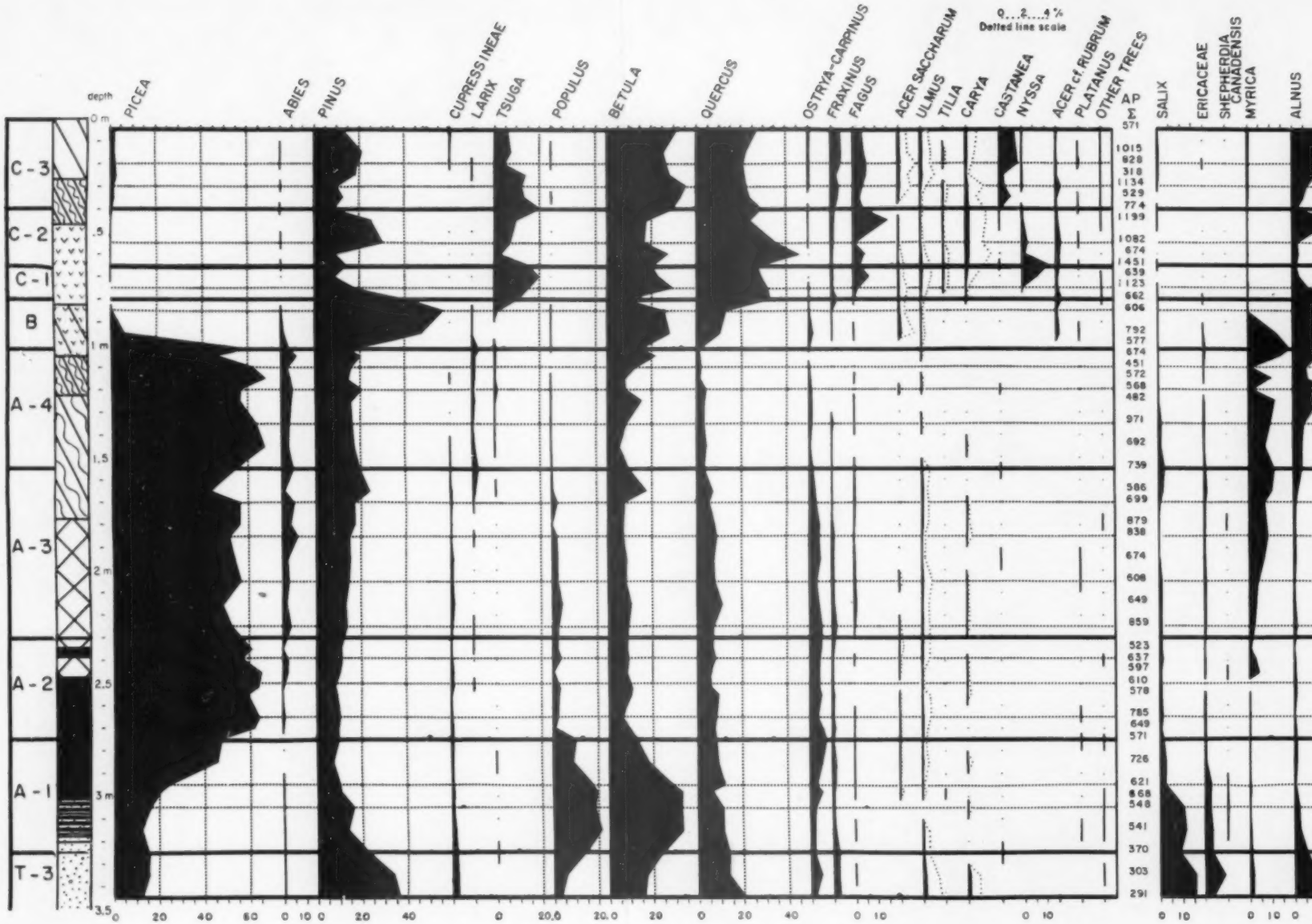
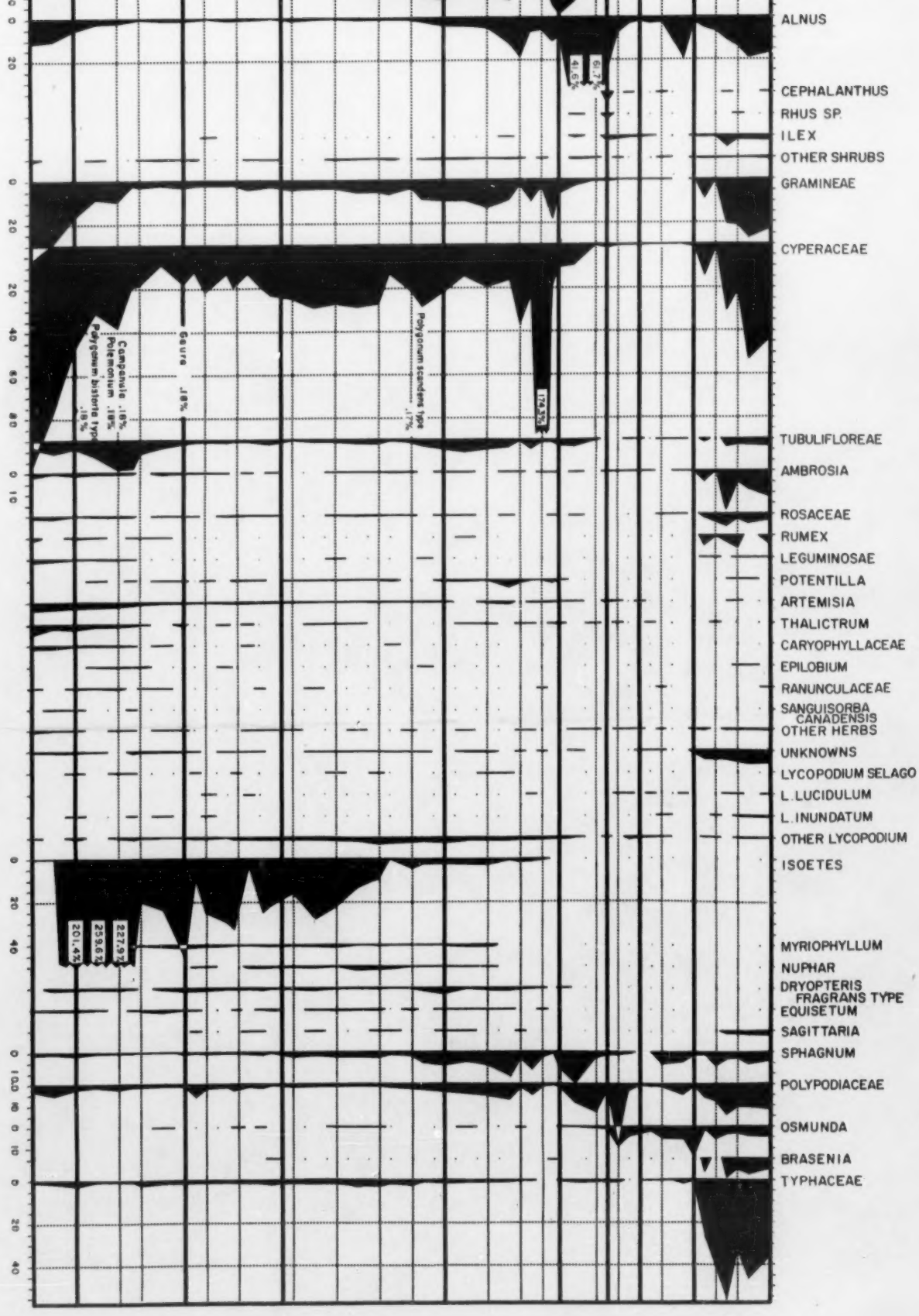


PLATE 4

Pollen diagram from Gould's Bog, Petersham, Mass. Percentage base Σ includes arboreal pollen only. Peat signatures are indicated in fig. 4.


ALNUS
CEPHALANTHUS
RHUS SP.
ILEX
OTHER SHRUBS
GRAMINEAE
CYPERACEAE
TUBULIFLOREAE
AMBROSIA
ROSACEAE
RUMEX
LEGUMINOSAE
POTENTILLA
ARTEMISIA
THALICTRUM
CARYOPHYLLACEAE
EPILOBIUM
RANUNCULACEAE
SANGUISORBA CANADENSIS
OTHER HERBS
UNKNOWNS
LYCOPODIUM SELAGO
L. LUCIDULUM
L. INUNDATUM
OTHER LYCOPODIUM
ISOETES
MYRIOPHYLLUM
NUPHAR
DRYOPTERIS FRAGRANS TYPE
EQUISETUM
SAGITTARIA
SPHAGNUM
POLYPODIACEAE
OSMUNDA
BRASENIA
TYPHACEAE
Campanula .16%
Pilemonium .18%
Polygonum bistorta type .18%
Geum .18%
Polygonum scandens type .17%
174.2%
4.6%
6.3%
201.4%
259.6%
227.9%

On the basis of these data, the 47 μ mode may represent jack pine (*P. banksiana*) and/or red pine (*P. resinosa*). The mode at 60 μ may represent a mixed population of white and pitch pine (*P. strobus* and *P. rigida*).

Spruce.—Size measurements of spruce pollen (see fig. 8) indicate that two types of spruce are present. Spruce with large pollen predominates in the lower spruce maximum (zone A-2), and spruce with small pollen predominates in the upper spruce maximum (zone A-4). Apparently both types are present in the intervening spruce minimum (zone A-3). Studies of the size frequency distribution of modern spruce pollen indicate that the average size of white spruce (*Picea glauca*) and red spruce (*P. rubens*) pollen is significantly larger than that of black spruce (*P. mariana*). The internal diameter of acetolyzed preparations of white spruce pollen (8 collections) averages 80.36 μ, red spruce (8 collections) 76.59 μ, and black spruce (3 collections) 62.17 μ (Cain, 1948; S. A. Cain and S. T. Andersen, personal communication).

As total grain length of the fossil spruce pollen, rather than internal diameter, was measured, the data cannot be compared directly with the measurements of modern pollen. However, as there are clearly two spruce types present, it seems safe to conclude that the spruce with large pollen in the lower spruce maximum is white or red spruce, and that the spruce with small pollen in the upper spruce maximum is black spruce. Spruce pollen could not be measured systematically in samples from the other sites because of the comparatively poor pollen preservation. However, the few measurements that were made of spruce pollen from Gould's Bog support the above conclusions. All spruce pollen from the maximum in zone C-3 at this site fell within the size range of black spruce.

INTERPRETATION OF THE POLLEN DIAGRAMS

The similarity of the three diagrams indicates that the vegetational sequence they record is regional in character. Probably a wide area in central Massachusetts had a similar vegetational history subsequent to the retreat of the ice. A knowledge of the vegetational sequence is of value in itself, but it is interesting to speculate on the factors which may have controlled it.

Interpretation of the diagrams is not easy. Because some plants produce pollen more abundantly than others, the frequency of the pollen of a genus or species in a pollen spectrum is not directly proportional to the frequency of that genus or species in the surrounding vegetation. The resulting difficulty in visualizing the vegetation represented by a pollen spectrum is illustrated by the uppermost sample from Gould's Bog. This sample is thought to date from late pre-colonial time. The tree pollen spectrum in the sample is dominated by birch and oak, with pine, chestnut, hemlock, ash, and beech of lesser importance. There is abundant pollen of non-arboreal plants, principally sedges, grasses, alder, and aquatic plants. By contrast, it is thought (Raup and Carlson, 1941) that the precolonial landscape was heavily forested, with oak, chestnut, and hickory on upland soils, and forests of birch, beech, maple, ash, elm, and hemlock on moist lowland sites. Pine was relatively unimportant. In the pollen spectrum, the pollen of herbs and aquatic plants is abundant because of their

proximity to the site of deposition. Of the trees, birch and pine, which produce abundant pollen, are overrepresented, while the maples and hickories, which produce less pollen, are underrepresented. It is well to keep these facts in mind in interpreting the diagrams. Changes in the relative abundance of the different pollen types, rather than the actual percentages, must be used to reconstruct the vegetation of the past.

Additional information is given by the presence or absence of the pollen of species which are known to occur only in certain types of environments. Unfortunately it has not been possible in most cases to make positive species identifications. Most American genera are so wide-ranging that their presence or absence yields little detailed ecological information. Furthermore one must assume, probably fallaciously, that the ecological requirements of species have remained stable since the last glaciation. An additional difficulty is presented by the irregular topography of the Petersham region. A wide variety of habitats are found there today, and presumably just as great a variety existed in the past. Thus the presence of the pollen of a species which occurs today in one type of environment does not preclude the possibility that plants typical of other types of environments were able to grow in the vicinity at the same time.

In the following discussion an attempt will be made to visualize the vegetation of the Petersham region in the past, and to inspect the edaphic and climatic factors that may have been responsible for the changes that are recorded. More than one interpretation of the diagrams is possible. The interpretation presented here, in the writer's opinion, fits best with the available data. Additional pollen diagrams from southern New England, more refined techniques of pollen identification, and a better understanding of the factors that control the present distribution of the species and genera involved are needed to test the validity of the interpretation.

Zones T-3, A-1, and A-2.—In the writer's opinion, zone T-3 represents an interval immediately following the retreat of the ice when a pioneer vegetation of shrubs and weedy herbs predominated. Frost-stirring, encouraged by the lack of forest cover (Goodlett, 1954), and landslides caused by the melting of buried ice, made the soil unstable. Strong winds deposited silt on the uplands. Under these conditions, only sheltered habitats on well-drained soils were suitable for the growth of trees. On these sites jack (or red) pine, white (or red) spruce, birch (perhaps shrub birch?), oak, and possibly other deciduous trees may have had a scattered distribution.

The evidence for this interpretation is the strong maximum of nonarboreal pollen at all three sites. It could be argued that the herbs were growing locally on the surface of bogs, but there is no indication from the sediments that the lakes were filling in with bog vegetation. The presence of many shade-intolerant herbs and shrubs is further evidence for an open, treeless vegetation.

Was the vegetation of zone T-3 a tundra? This question is hard to answer, because of the variety of vegetational types that are included under this heading. In a floristic sense, it was not a tundra. Many genera which never grow in tundra regions were found in this zone, and no species that are

characteristic of tundra regions alone were identified positively. Although the vegetation of zone T-3 may have resembled a tundra in aspect, the name tundra would be misleading. "Tundra" implies to most people a vegetation which is treeless because the climate is too cold to allow the growth of trees. There is as yet no conclusive evidence that the climate of T-3 was arctic. Only the presence of *Saxifraga oppositifolia*, which was tentatively identified argues for a tundra climate. The other species with a northerly distribution that were found in T-3 and the succeeding zones, such as *Polygonum* cf. *bistortoides*, *Lycopodium selago*, *Selaginella selaginoides*, *Sanguisorba canadensis*, *Sheperdia canadensis*, and *Hippuris* cf. *vulgaris*, indicate boreal, or possibly subarctic climates, rather than arctic ones. And there are many genera which, at least in the northeast, indicate temperate climates. Among the herbs there are *Polemonium*, *Ambrosia*, and the Chenopodiaceae, and among the trees oak, hop-hornbeam, and several other deciduous genera. Surely indicators of temperate climate should be given as much consideration as indicators of tundra.

The higher percentages of tree pollen, principally pine, in the outwash sands underlying Tom Swamp and Pleasant St. Bog are somewhat difficult to explain. It is possible that the sands were deposited by glacial melt-water in the short interval before vegetation became established in the area. Under these circumstances the only pollen deposited was windblown pollen from vegetation to the south. Several arguments support this hypothesis. First, the absolute pollen frequency in the sand at these two sites was extremely low, and special extraction techniques were necessary. The percentage weight lost on ignition was also extremely low. Second, the pollen spectra from the sand are dominated by pine, which produces abundant, buoyant pollen which is easily transported by wind. Third, studies in Lapland (Aario, 1944) have demonstrated the surprising fact that surface samples from the far north have higher pine pollen percentages than surface samples from treeless areas near the forest limit. This is because the herbaceous flora in very cold, dry tundra regions is scanty, and does not produce pollen in abundance. Therefore the ratio of windblown pollen to locally produced pollen increases with increasing distance from the forest limit. For these reasons the pollen spectra in the outwash sands underlying zone T-3 at Tom Swamp and Pleasant St. Bog are not considered reliable indicators of vegetation.

Zone A-1 records the development of a continuous forest cover. Whereas spruce dominates at the forest limit in the eastern arctic at the present time, balsam poplar formed the tree-line of the advancing forest in central Massachusetts during late-glacial time. Its light, easily-transported seed (Halliday and Brown, 1943) and its ability to pioneer on disturbed mineral soil may have been responsible for its success. Birch grew together with the poplar. *Betula glandulosa*, a northern shrub birch, may have grown as an understory in the open forest; or gray birch (*B. populifolia*), a temperate tree birch which is able to grow on disturbed soils, may have been present. Herbs and shrubs, apparently shaded by poplar, began to decline in frequency; a few may have persisted in the more exposed habitats.

The poplar was soon succeeded by a spruce forest. Size measurements of the pollen indicate the presence of white and/or red spruce. High percentages

of spruce at Tom Swamp may indicate that the outwash surrounding this site was a particularly favorable habitat for spruce. Zones T-3, A-1, and A-2 appear to record a succession from herbaceous vegetation to poplar forest to spruce, similar to a succession which occurs today on flood-plains in the Mackenzie region (Raup, 1935), and on the subarctic prairies (Raup, 1941). In marked contrast to northern Europe, where treeless vegetation persisted for a long period after the retreat of the ice, the forest appears to have advanced in central Massachusetts as fast as soil stabilization and migration speed would allow. The treeless zone along the edge of the ice sheet was probably narrow. Denny (1956b) has shown that in Pennsylvania a zone only ten miles wide south of the Cary drift underwent severe periglacial frost action. Similarly, in New England, intense frost action was probably confined to a zone only a few miles wide south of the retreating ice.

Because soils and migration speed, rather than climatic changes, appear to have controlled the vegetation in zones T-3, A-1, and A-2, these zones probably are not strictly contemporaneous with T-3, A-1, and A-2 in Connecticut (Leopold, 1955, 1956b). Gyttja from zones A-1 and A-2 in Connecticut has been dated by radiocarbon analysis at 12,700 ± 280 and 13,550 ± 460 years BP (Leopold, 1956b), and 14,790 ± 160 and 13,290 ± 120 years BP (Barendsen, et al., 1957). These dates imply that zone A-2 in Massachusetts is of Two Creeks age.

Zones A-3 and A-4.—A poplar log from zone A-3 at Pleasant St. Bog has been dated by radiocarbon analysis at 10,700 ± 800 and 10,800 ± 280 years BP (see Description of Sites). The dates indicate that zone A-3 is contemporaneous with either late Two Creeks or early Valders deposits in the midwestern sequence. Zones A-3 and A-4 are difficult to interpret. A wide variety of species and genera, many of them plants not usually found together today, occur in these zones. There appear to be two possible interpretations of the sequence. One interpretation, following Leopold's correlation (Leopold, 1956b), assumes that all the pollen is from plants that grew in the vicinity. The other interpretation assumes that some of the pollen in zone A-3 and the preceding zones has been redeposited from older deposits. Following the former hypothesis, we find that the vegetation of zone A-3 was a mixed spruce and deciduous forest. Spruce and an occasional fir may have grown on the uplands while oak, hop-hornbeam, elm, sugar maple, and beech grew in sheltered well-drained habitats. Ground juniper or red cedar (or arbor vitae?) was common. Shrub birch may have grown on swampy sites, or gray birch and a few other tree birches may have grown together with the other deciduous trees. A similar mixing of boreal and deciduous trees south of the ice during the glacial maximum has been postulated by Drury (1956). Certainly the irregular topography and mosaic distribution of till and outwash deposits in the Petersham region would have provided a variety of habitats. Today mixed forests can be found at altitudes of 1600 ft and higher in the Berkshire mountains 80 km (50 mi) west of Petersham, and in southern New Hampshire 100 km (65 mi) to the north. In fact a type of beech, the "northern gray beech", is reported to occur in a pure genetic state only when growing together with spruce or fir (Camp, 1950). The vegetation of zone A-3 is pecu-

liar because of the absence of hemlock (*Tsuga*), white pine (*Pinus strobus*), and red maple (*Acer rubrum*), which are common in mixed forests today, and in the abundance of hop-hornbeam (or ironwood?). However, the climatic and edaphic conditions of zone A-3 are probably not duplicated anywhere at the present time.

The pollen size frequency data indicate that jack and/or red pine are present in this zone. The maximum of pine along the A-3—A-4 border is hard to explain. Possibly a dry and cooler climate favored an expansion of pine.

A maximum of *Myrica* (and birch?) at Tom Swamp and Pleasant St. Bog, and rising percentages of sedges, *Myrica*, and grasses (and birch?) at Gould's Bog indicate the development of local bog vegetation. Apparently the lakes had begun to fill in. At the first two sites the *Myrica* maximum is followed by a rise of *Isoetes* and sphagnum. At Gould's Bog growth of the bog mat continued, and the pond was completely filled in with vegetation in late A-4 time.

Zone A-4 is marked by an expansion of black spruce and a decrease of deciduous trees. Today black spruce occurs as an upland tree on north-facing slopes in the boreal forest (Raup, 1947); perhaps it is favored there by lower temperatures, greater available moisture, or a combination of the two. We may postulate a decrease in temperature and an increase in precipitation to explain the success of this species in zone A-4. The rise of larch and hemlock, and the presence of pine and birch of larger pollen size, perhaps representing white pine and yellow birch, also may be interpreted as evidence of increased precipitation. Following this interpretation, zone A-4 is correlated with an advance of ice contemporaneous with the Valders advance in Wisconsin. The glacial border must have been far to the north; apparently the ice itself did not influence the climate of central Massachusetts. The landscape remained heavily forested; the climatic oscillation associated with the ice advance merely resulted in a change of percentage composition among the forest species.

However, there is some evidence against the interpretation that has just been presented. The pollen size measurements indicate that the pine of zone A-3 may be jack pine. If this is the case, the presence of white or pitch pine in zone A-4 (see fig. 7) may indicate a warmer climate. Similarly, if the birch in zone A-3 is shrub birch, the increase in frequency of large birch pollen in A-4 (see fig. 6) means an increase of warmth-demanding tree birches. We can interpret zone A-3 as a cold period, and A-4 as a warmer one, if we assume that some of the pollen has been rebedded from older deposits. If the tills of the Petersham region contain pollen from interglacial deposits overridden by the ice sheet, the pollen, together with particles of inorganic matter, would be carried into the lakes during periods of intense solifluction. There it would be rebedded with pollen from the local vegetation. Many deposits in Europe are thus contaminated with Tertiary or interglacial pollen (Iversen, 1936). In general, the amount of secondarily deposited pollen varies directly with the clay content of the sediment (Iversen, 1947; Krog, 1954); in purely organogenic deposits rebedded pollen may be absent entirely (Iversen, 1947). If the entire spruce zone is contaminated with rebedded pollen of temperate species, the zone with a maximum of pollen of temperate trees, A-3, represents a

period of intense solifluction. Zone A-2 would thus represent spruce forests established in Two Creeks time. Zone A-3 would represent the Valders stadium, with unstable soils supporting a park-tundra of spruce and jack pine, arctic juniper, shrub birch and herbs. Zone A-4 would represent the reestablishment of forests in early postglacial time. This interpretation would yield a vegetational sequence similar to that found in southern Germany (Lang, 1952).

In the writer's opinion, the assumptions necessary for this interpretation are not justified by the evidence. The ignition loss curves (figs. 3 and 4) do not show an increase in inorganic matter in zone A-3 which can be correlated with the supposed increase of rebedded pollen. Furthermore, if the deciduous tree pollen is rebedded, the ratio of different types should be the same throughout the lower part of the diagrams. This is not the case, especially at Gould's Bog. If pollen of temperate deciduous trees is considered rebedded, then pollen of temperate herbs must also be included in this category. Subtraction of all temperate herbs would considerably lower the maximum of NAP in A-3 which is part of the evidence for a return of park-tundra. The loss on ignition data do show that zones T-3 and A-1 at all three sites are considerably less organic than the succeeding zones. There is a strong possibility that some of the pollen in these zones is rebedded. At Gould's Bog, the percentages of deciduous trees in these zones are somewhat higher than at the other sites. Basswood, sycamore, hackberry, and black walnut, which were not found below the pine zone at Tom Swamp and Pleasant St. Bog, although large numbers of pollen were counted, occur in zone A-1 at Gould's Bog. Perhaps at this site there is a very small percentage of rebedded pollen. However, there is as yet no conclusive evidence to prove the validity of this hypothesis.

Zone B-1.—Zone B-1 records the sudden decline of spruce. Forests of red and/or jack pine and birch were widespread. Oak and other deciduous trees, although still not abundant, had a scattered distribution. Alder, perhaps together with larch, grew on the shores of the lakes, and at Gould's Bog, replaced *Myrica* as the dominant bog shrub. The low percentages of deciduous tree pollen suggest that the climate was still cool. The rapid decline of spruce may have been caused by a change from the moist conditions of zone A-4 to a dry continental climate.

Zone B-2.—In B-2 time there were extensive forests of pine and oak. Size measurements of pollen (see fig. 7) indicate that white and pitch pine, rather than jack and/or red pine, were the dominant pine species. Forests of pitch pine and oak are common at present on sand plains in central Massachusetts (Bromley, 1935). Under conditions somewhat warmer and drier than the present, they may have occurred on all the well-drained soils, while hemlock and other deciduous trees, such as hop-hornbeam or ironwood, ash, elm, sugar maple, and red maple occupied the wetter sites. Dry climate is indicated by the change at all three sites from peat deposited under more hydrophytic conditions to peat deposited under less hydrophytic conditions. The lower boundary of zone B-2 is considered here to represent the beginning of the

hypsithermal interval (Deevey and Flint, 1957), a period when prevailing temperatures were higher than they are today.

Zone C-1.—In C-1 time the climate appears to have been more moist. Pine became less common, and oak, perhaps a different species than in zone B-2, increased in frequency. Hemlock, elm, sugar maple, and basswood were abundant. Warmth-demanding trees, such as hackberry and sycamore, now only occasional in central Massachusetts, occurred in low frequencies. Beech became more common, reaching a maximum along the C-1—C-2 zone boundary.

Zone C-2.—Apparently zone C-2 represents the warmest part of postglacial time. The climate was also fairly dry, as indicated by the minimum of hemlock and elm, and by the terrestrial peat deposited at all three sites. The forests were dominated first by oak, and then by oak and pine. Hickory was slightly more abundant than it is today. Sugar maple was at a minimum.

Low water level at Gould's Bog during C-2 time is implied by poor pollen preservation and highly humified peat. Apparently a bog forest of black gum, red maple, and possibly birch in early C-2 was replaced in late C-2 time by alder. Perhaps beech was also growing on the bog. The beech maximum of late C-2, although characteristic of this horizon in New England (Deevey, 1943), seems anomalous in that elm, sugar maple, and hemlock, which occur together with beech in the other pollen zones, are almost absent. It seems likely that the maximum represents a different beech type, possibly the "white beech" which is reported to be common in swamps further south (Camp, 1950). The successive maxima of beech in New England pollen diagrams have been attributed to waves of migration (Deevey, 1949). Perhaps the maximum of late C-2 time is due to the influx from the south of new genetic material which enabled the beech population to expand into different habitats.

Zone C-3.—Zone C-3 records the most recent stage in the history of the precolonial forests. Oak and hickory declined in frequency, and hemlock, elm, sugar maple, and birch became abundant. These changes may indicate a cooler and more humid climate. Chestnut and ash for the first time became important members of the forest complex. In the upper part of the zone, hemlock declines and pine rises to a maximum. The rise of spruce, noted in Connecticut (Deevey, 1943), probably represents the occurrence of black spruce on the bog surface. A more humid climate also is indicated by the change in the nonarboreal bog vegetation. The Typhaceae and many bog herbs and shrubs suddenly increase in frequency. The peat is fibrous and less humified, and pollen preservation is far better than in the underlying zone. This change from a dry, wooded swale to a wet, sedge and cattail bog at the beginning of zone C-3 is analogous to the sudden resumption of bog growth in northwestern Europe at the beginning of Sub-atlantic time. Recurrence horizon III, the most pronounced of the *Grenzhorizonten*, which marks the lower border of the Subatlantic zone in Europe, has been dated at about 600 B.C. (Granlund, 1932). The stratigraphy of Gould's Bog is evidence that a sudden change from warm-dry to cool-moist conditions occurred at about the same time in northeastern North America, as well. Probably the end of the hypsithermal interval (the

end of the Sub-boreal zone, the end of zone C-2), occurred simultaneously in both continents at about 600 B.C.

General remarks.—It is interesting and important in considering the concept of the vegetational "climax" to realize that the pre-colonial forest of Petersham, which was rich in chestnut and ash, differentiated only recently. We may well question the idea that plant associations are continuous in time. Rather, it appears from the diagrams that each species (or ecotype) has reacted independently to changes in climate and soil. The concept of vegetational succession should also be considered in the light of this historical evidence. The history of Gould's Bog has been far more complex than its present aspect would indicate. If "the stages in the present horizontal sequence from shallow water to marginal forest become arranged in a vertical sequence as the bottom of the lake is built up" (Weaver and Clements, 1938, p. 66), we should expect only aquatic sediments below the present bog surface. Instead, there is recorded the invasion of open water by a floating sedge mat, the growth of first a *Myrica* and then an alder (and larch?) thicket, the development of a hardwood swale forest, and the subsequent invasion of the swale by hydrophytic vegetation. It is true that under certain circumstances the present vegetational zonation around the shores of a bog may represent successional stages. This is the case at the north and south margins of Tom Swamp, where the encroachment of vegetation into open water during the last 15 years is recorded by aerial photographs. However, Gould's Bog and the deeper parts of Tom Swamp represent far more typical cases of bog succession. Here the development of the bog has taken many thousands of years. During this time the water table, the surrounding vegetation, and the soils have undergone many changes. The nature of the bog vegetation, and the direction and speed of plant succession appear to have been under climatic control.

The pollen diagrams presented here are easily correlated with diagrams from Connecticut (Deevey, 1939, 1943; Leopold, 1955, 1956b) and southern New Hampshire (Krauss and Kent, 1944). However, the early part of the sequence, zones T-3—B-1, is strikingly different from the early sequences found in northern Maine (Deevey, 1951), Nova Scotia (Livingstone and Livingstone, 1958), Michigan (Andersen, 1954), and Europe (Firbas, 1950; Donner, 1951; van der Hammen, 1951; Lang, 1952; Iversen, 1954; Godwin, 1956). In these areas there is evidence of persistence of tundra for some time after the retreat of the ice, and, where forests became established, a return of tundra or park-tundra during Valders (Younger Dryas) time. Although it seems strange that the early sequence in southern New England is anomalous, should we expect it to be similar to late-glacial sequences elsewhere? In contrast to Europe, there are no east-west mountain chains to serve as barriers to migration. Boreal and temperate species were available for recolonization (Drury, 1956). As soon as the climate and soils were favorable for the growth of trees, forests were established. The Alps served as a barrier to migration in Europe, and, as a result, in some areas slow migration speed, rather than unfavorable climate, may have hindered the development of boreal forest (van der Hammen, 1951; Iversen, 1954). Furthermore, when sea levels were low, central Massachusetts was far from the ocean; the climate may have been

even more continental than it is now. There were no large proglacial lakes, such as the Baltic ice-lake or ancestral Great Lakes, to exert an oceanic influence. A continental climate at the comparatively low latitude of Massachusetts (42°N) would have resulted in high summer temperatures, permitting the growth of many species that could not have survived during the same period in Europe (cf. Firbas, 1950). Rather than attempting to compare our late-glacial sequence with that of northwestern Europe, we should look to continental areas, such as eastern Europe, for a similar vegetational history.

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THE NEPHELINE-KALSILITE SYSTEM
II. PHASE RELATIONS*

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ABSTRACT. Phase equilibrium studies in the system $NaAlSiO_4$—$KAlSiO_4$ at subsolidus temperatures have demonstrated that a solvus, or miscibility gap, is present and in a general way it is similar to the solvus between albite and orthoclase. At low temperatures the miscibility gap extends in composition from $Na_3KAl_4Si_4O_{16}$ to $KAlSiO_4$.

The stable modifications of $KAlSiO_4$ were found to be kalsilite and a high temperature orthorhombic modification while kaliophilite is believed to be a metastable phase at all temperatures at atmospheric pressure. $NaAlSiO_4$ has a high temperature form stable between 850°C and 1250°C. A new phase, tetrakalsilite, was produced in mixtures heated at high temperatures in compositions near $K_2NaAl_3Si_4O_{16}$.

Evidence is presented to show that $Na_3KAl_4Si_4O_{16}$ is a compound (nepheline) and not merely a single composition in a solid solution series.

INTRODUCTION

As experimental studies on the $NaAlSi_3O_8$—$KAlSi_3O_8$—SiO_2—H_2O system drew to a close (Tuttle and Bowen, 1958) x-ray powder diffraction studies were carried out on the crystalline phases in the $NaAlSiO_4$—$KAlSiO_4$ system preparatory to extending the liquidus studies into this part of the $NaAlSiO_4$—$KAlSiO_4$—SiO_2—H_2O system. Preliminary x-ray studies on these mixtures, previously prepared by Bowen (1917) and by Bowen and Schairer (1935), disclosed several interesting and puzzling features that made it advisable to undertake a detailed study of the subsolidus relations in the system $NaAlSiO_4$—$KAlSiO_4$ before proceeding into the compositions with higher silica.

The x-ray properties of the phases encountered in the system are described in a previous publication (Smith and Tuttle, 1957).

EXPERIMENTAL METHODS

We are indebted to Drs. Bowen and Schairer for permission to use the mixtures which were prepared for their study of this system (Bowen, 1917 and Bowen and Schairer, 1935). X-ray studies of crystallized mixtures in the composition range Ne_{100} to [1]Ne_{30} indicated that they consist of a single-phase solid solution. Preparations having compositions in the range Ne_{30} to Ne_0 gave x-ray patterns that indicated the presence of two or more phases.

Preliminary studies on the x-ray powder diffraction patterns of the solid solutions in the composition range Ne_{100} to Ne_{30} suggested that the reflections shifted progressively with changes in composition of the nepheline solid solutions. This change in d-spacing as a function of composition was used to estimate the composition of the solid solutions in solidus and subsolidus studies. The (21.0) and (20.2) spacings were found to be suitable for estimating compositions and a determinative graph was prepared (fig. 1). In practice, silicon powder is mixed with the unknown and the angles between the (111) reflection of silicon and the (20.2) reflections of the solid solutions are measured.

* M. I. Contribution No. 57-35.

[1] Ne = $NaAlSiO_4$, Ks = $KAlSiO_4$, Ne_{30} = 30 wt. percent $NaAlSiO_4$, 70 wt. percent $KAlSiO_4$, etc. Although the ideal composition of nepheline is believed to be $Ne_{72.9}$ ($Na_3KAl_4Si_4O_{16}$) solid solutions in the compositional range Ne_{30}-Ne_{100} will be referred to as nepheline solid solutions (see page 583).

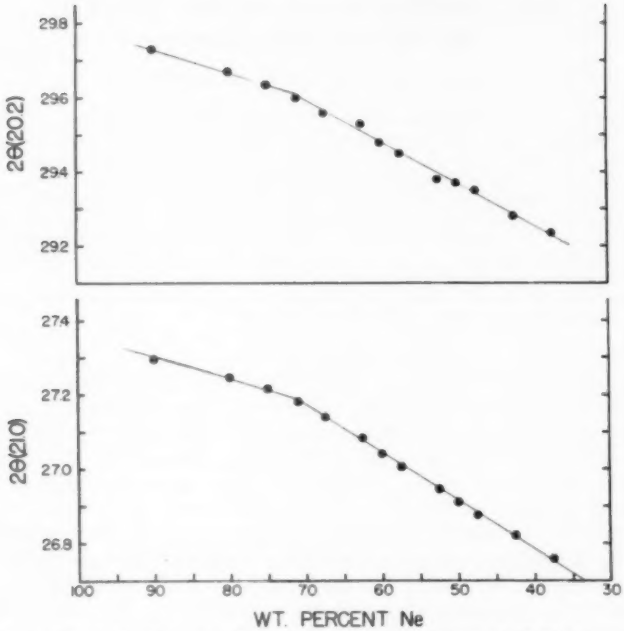

Fig. 1. Graph showing the change in two x-ray reflections (2θCuKα) in nepheline solid solutions as a function of composition (calibrated with silicon (111) spacing 2θCuKα = 28.466°). These reflections can be used to estimate the composition of natural nephelines (see Smith and Sahama, 1954).

Mixtures having compositions between $Ne_{37.5}$ and Ne_0 were not single phases. The inhomogeneity results in part from 1) the presence of small amounts of leucite as a result of loss of alkalies during preparation of the mixture, 2) polymorphism, and 3) limited solid solutions in the various polymorphic modifications. It is not unusual to obtain three or four phases in the low temperature runs in these compositions, and in some experiments the x-ray patterns were so complicated that the various phases could not be definitely identified.

Kalsilite will take up to approximately 20 percent (page 581) $NaAlSiO_4$ in solid solution with appreciable shift in lattice constants, and the diffraction angles for the (10.1) and (10.2) planes have been found useful in estimating compositional variations in synthetic material. These reflections are strong and can easily be distinguished from those of nepheline solid solutions (Smith and Tuttle, 1957).

Four methods were used to study the subsolidus phase relations: 1) dry quenching (Shepherd and Rankin, 1909), 2) hydrothermal quenching (Tuttle, 1948), 3) differential thermal analysis, and 4) high temperature x-ray studies. The solvus was first located by hydrothermal quenching experiments at low

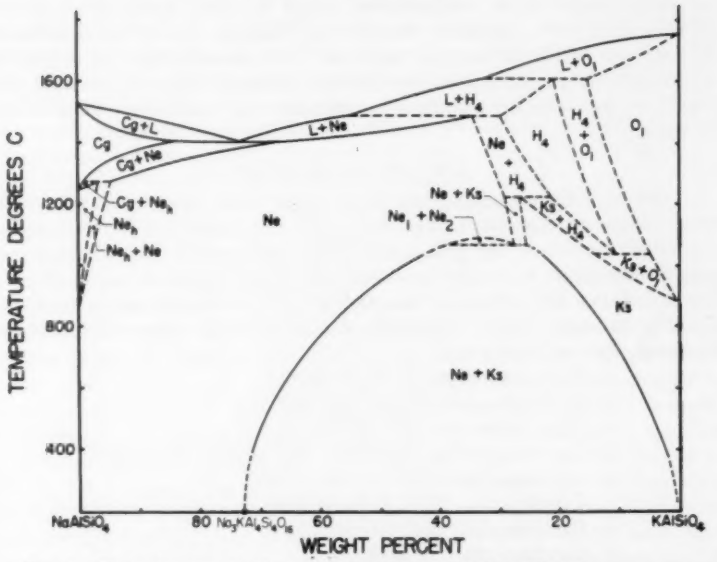

Fig. 2. Phase diagram for the system $NaAlSiO_4$—$KAlSiO_4$. This diagram represents an attempt to assign a stability field for the new phase tetrakalsilite (H_4). The dashed lines are inferred phase boundaries. Cg = carnegieite, L = liquid, Ne_h = high temperature nepheline, Ne = low temperature nepheline, O_1 = orthorhombic $KAlSiO_4$, Ks = kalsilite, and H_4 = tetrakalsilite.

temperatures. Later studies by dry quenching and high temperature x-ray diffraction methods served to fix the miscibility gap as shown in figure 2.[2] Differential thermal analysis was used to locate the high-low inversion in nepheline. The inversion kalsilite ⇄ orthorhombic $KAlSiO_4$ was fixed by hydrothermal quenching methods using cold seal pressure vessels (Tuttle, 1949). This inversion is sluggish and the high temperature form (orthorhombic $KAlSiO_4$) can be preserved at room temperature by rapid cooling.

Some of the preparations used in this study deviate from the stoichiometric proportions in the system $NaAlSiO_4$—$KAlSiO_4$ because potassium and sodium oxide are lost by volatilization during the preparation of the mixtures. This deviation is believed to be small in preparations rich in nepheline, but may be considerable in compositions rich in kalsilite (see Bowen, 1912, p. 552 and 1917, p. 116). In the compositional range Ne_{100}– $Ne_{37.5}$ only a single phase could be identified in x-ray powder diffraction patterns, and the compositional changes due to volatilization are therefore probably not large. These compositions did not develop leucite during the present study despite the fact that runs were made at 1520°C for as long as six hours. In contrast,

[2] Pressure will alter the position of the solvus and theoretically the dry and wet determinations should give slightly different values; however, since 1000 Kg/cm² is the highest pressure used, it is probable that the effect of pressure is not greater than the error of the measurements.

the preparations in the compositional range Ne_0–Ne_{30} were inhomogeneous initially and often contained leucite after heating dry or hydrothermally. The amount of leucite was not large and it is probable that our subsolidus studies are not greatly in error by this loss of potash; however, the final products were not analyzed and, as a consequence, this uncertainty must be kept in mind.

THE CRYSTALLINE PHASES

Ten crystalline phases have been prepared in mixtures having initial compositions in this system. Two were discovered during this study, and, as will be shown below, one ($Na_3KAl_4Si_4O_{16}$) is believed to be an unique compound although it is usually considered to represent a single composition in the nepheline solid solution series. At least four of the ten phases are known to occur naturally and it is probable that some of the others will eventually be found. The ten phases are:

1. High carnegieite (Cg). $NaAlSiO_4$
2. Low carnegieite (Cg). $NaAlSiO_4$
3. High temperature $NaAlSiO_4$ (Ne_h).
4. Low temperature $NaAlSiO_4$, Nepheline[3] ($Na_3KAl_4Si_4O_{16}$) and nepheline solid solutions (Ne)[4]
5. Tetrakalsilite (H_4). $K_3NaAl_4Si_4O_{16}$
6. Orthorhombic $KAlSiO_4$ (O_1).
7. Kalsilite[3] (Ks). $KAlSiO_4$
8. Kaliophilite[3] (Kp). $KAlSiO_4$
9. Disordered kaliophilite (Kp). $KAlSiO_4$
10. Orthorhombic phase (O_2) having *a* and *c* parameters the same as orthorhombic $KAlSiO_4$ but *b* is two-thirds smaller. Composition unknown.

Low temperature $NaAlSiO_4$, nepheline, tetrakalsilite, kalsilite, and kaliophilite are hexagonal; high carnegieite is cubic at elevated temperatures and inverts metastably to a low symmetry phase at 690°C. All other phases have orthorhombic symmetry.

X-ray properties of the various phases are given in an earlier publication (Smith and Tuttle, 1957).

EXPERIMENTAL RESULTS

The Solvus.—Experience gained in the study of phase relations in the system $NaAlSi_3O_8$—$KAlSi_3O_8$—H_2O (Bowen and Tuttle, 1950) suggested that the complete solid solution in the $NaAlSiO_4$—$KAlSiO_4$ system reported by Bowen (1917) might well give way to a miscibility gap (solvus) at low temperatures. Accordingly, our earliest experiments were designed to locate a solvus, if present. X-ray studies of the mixtures prepared in the dry way by Bowen (1917) and by Bowen and Schairer (1935) indicated that solid solution was indeed extensive, and only those compositions rich in kalsilite gave

[3] Phases occurring as natural minerals.

[4] $NaAlSiO_4$ and $Na_3KAl_4Si_4O_{16}$ are here distinguished as separate compounds but as an unbroken series of solid solutions extends between these compounds, only one low temperature nepheline phase exists.

x-ray patterns which suggested the possible presence of two phases. The first convincing evidence of a miscibility gap was discovered in mixtures heated in the presence of water vapor under pressure. Some of the preparations which were *initially homogeneous single phases broke into two phases when heated at 800°C* and 1000 Kg/cm² water vapor pressure. Subsequent experiments at different temperatures gave consistent evidence that an extensive solvus exists at temperatures below 1050°C. Results of this part of our study are shown in figure 2 and individual experiments are presented in tables 1 and 2. Solid lines in figure 2 indicate the phase relations which have been directly and reproducibly determined whereas dashed lines represent our interpretation of the relations where data are incomplete or inconsistent.

Experimental results on the mixtures Ne_{30}, Ne_{20}, and Ne_{10} are in part inconsistent and the phase relations between the top of the solvus and the solidus are schematic and represent an attempt to assign a stability field for the new phase tetrakalsilite and at the same time allow for a break in the nepheline to kalsilite solid solutions. Experimental difficulties encountered in the region just above the solvus are probably the result of 1) failure to reach equlibrium, 2) compositional uncertainties, and 3) failure to quench the high temperature phases or assemblages. For example, the Ne_{20} mixture heated at 1200°C for 17 hours was largely tetrakalsilite on quenching whereas after 168 hours it had changed to kalsilite plus a little tetrakalsilite; at 1400°C the same mixture was largely kalsilite after six hours and after 17 hours had changed to tetrakalsilite with a little kalsilite. It is known that runs carried out for long times, i.e., 168 hours, at these high temperatures lose alkalies by volatilization and we were faced with the dilemma: Should we make longer runs and risk severe compositional changes, or should we accept the indications of shorter runs? We chose the latter course and therefore, except for the solvus below 1050°C and the inversions in $KAlSiO_4$ and $NaAlSiO_4$, our results should be considered a reconnaissance and not a final detailed study of the subsolidus phase relations.

TABLE 1

Experimental Data

Abbreviations: O_1 and O_2 = orthorhombic $KAlSiO_4$; Sk = synthetic kaliophilite; Lc = leucite, Ks = kalsilite; Ne = nepheline solid solution; H_4 = tetrakalsilite

Initial Material		Heating conditions			Results
Composition Ne wt.%	Condition	Temp. °C	P_{H_2O} Kg/cm²	Time hours	
0	?*	1550	0	?	O_1
0	?*	1300	0 (KWO₄ flux)	48	O_1
0	$K_2Si_2O_5.\gamma Al_2O_3$	1100	500	1	O_1
0	?*	1075	0	?	Sk
0	$K_2Si_2O_5 + \gamma Al_2O_3$	1000	500	1	O_1
0	$K_2Si_2O_5 + \gamma Al_2O_3$	950	500	1½	O_1
0	Ks grown at 800°C	900	500	16	O_1 + little Lc
0	$K_2Si_2O_5 + \gamma Al_2O_3$	900	500	2	O_1

TABLE 1 (Continued)

Initial Material		Heating conditions			Results
Composition Ne wt.%	Condition	Temp. °C	P_{H_2O} Kg/cm²	Time hours	
0	$K_2Si_2O_5 + \gamma Al_2O_3$	855	500	16	complicated, possible Sk
0	$K_2Si_2O_5 + \gamma Al_2O_3 + 5\%$ excess Al_2O_3	840	500	5½	Kp + O_1 + little Lc + Ks ?
0	$K_2Si_2O_5 + \gamma Al_2O_3$	825	500	1½	Ks + Sk
0	O_1 made at 950° and 1100°C.	800	500	16	Ks + little Lc
0	$K_2Si_2O_5$—corundum	800	500	24	Ks + very weak Lc
0	$K_2Si_2O_5 + \gamma Al_2O_3$	800	500	72	central charge Ks + very weak Lc, buffer Ks + weak Lc
0	$K_2Si_2O_5 + \gamma Al_2O_3$	800	500	3¾	Ks + weak Lc
10	O_1	1520	0	6	O_1
10	O_1	1400	0	6	O_1 only
10	O_1	1200	0	168	O_1 + Ks
10	O_1	1200	0	72	O_1 + Ks + little Lc
10	O_1	1100	500	2	Very complicated probably some Sk, possibly Ks, little Lc.
10	O_1	1000	500	24	Very complicated possibly some Sk
10	O_1	800	500	16	Ks + little Lc
10	O_1	505	500	72	Ks (Ne_4) + little Ne(Ne_{12}) + little Lc.
20	Glass + O_2	1540	0	4	O_2 only
20	Glass + O_2	1520	0	6	O_2 + Ks(?) + little Lc
20	Glass + O_2	1400	0	6	Ks + little H_4
20	Glass + O_2	1400	0	17	H_4 + little Ks
20	Glass + O_2	1200	0	17	H_4 + little Ks
20	Glass + O_2	1200	0	72	Ks + H_4
20	Glass + O_2	1200	0	168	Ks + H_4
20	Glass + O_2	1100	500	2	Ks + H_4 + Lc ?
20	Glass + O_2	1000	500	6	Ks + H_4 + little Lc.
20	Glass + O_2	1000	500	2	Ks + H_4 + Lc ?
20	Glass + O_2	880	500	72	Ks(Ne_{12}) + little Ne(Ne_{20}) + little Lc
20	Glass + O_2	800	500	16	Ks(Ne_{12}) + little Ne(Ne_{20}) + Lc
30	Ne + H_4	1520	0	6	H_4 only
30	Ne + H_4	1400	0	6	Ks + Ne + little H_4
30	Ne + H_4	1400	0	17	H_4 + Ne
30	Ne + H_4	1200	0	17	H_4 + Ne
30	Ne + H_4	1200	0	72	Ne($Ne_{20.5}$) + Ks(Ne_7) + little H_4
30	Ne + H_4	1200	0	168	Ne(Ne_{21}) + Ks
37.5	Glass	1250	0	48	Ne
37.5	Glass	1150	500	1	Ne(Ne_{16}) + Ks(Ne_{11})
37.5	Glass	1100	0	96	Ne
37.5	Cryst. 1250° 16 hrs. 650°C dry	1100	0	20	Ne only
37.5	Glass	1100	500	1	Ne(Ne_{15}) + Ks (Ne_9)

TABLE 1 (Continued)

Initial Material		Heating conditions			Results
Composition Ne wt.%	Condition	Temp. °C	P_{H_2O} Kg/cm²	Time hours	
37.5	Glass	1050	0	96	Ne + Ks
37.5	Glass	1050	500	1	$Ne(Ne_{36})$ + $Ks(Ne_{18})$
37.5	Glass	1000	0	144	Ne + Ks
37.5	cryst. 1250° held 120 hrs. 650°C dry	1000	0	17	Ne + Ks
37.5	144 hrs. dry, 1000°C from glass	1000	500	3	$Ne(Ne_{36})$ + $Ks(Ne_{18})$
37.5	Glass	1000	1000	0.5	$Ne(Ne_{36})$ + $Ks(Ne_{18})$
37.5	cryst. 1200°C to Ne	1000	0	436	$Ne(Ne_{36.5})$ + Ks?
37.5	Glass	1000	1000	0.5	$Ne(Ne_{36})$ + $Ks(Ne_{18})$
37.5	Glass	950	0	144	Ne + Ks
37.5	Glass	900	0	700	poorly crystalline
37.5	Glass	800	1000	16	$Ne(Ne_{36})$ + $Ks(Ne_{18})$
37.5	Glass	800	1000	1.5	$Ne(Ne_{36}$ + $Ks(Ne_{11})$
37.5	Glass	800	1000	1.25	$Ne(Ne_{36.5})$ + $Ks(Ne_{18})$
37.5	Glass	600	500	96	$Ne(Ne_{37.5})$ + $Ks(Ne_9)$
37.5	Glass	505	1000	96	$Ne(Ne_{70})$ + $Ks(Ne_4)$
42.5	Glass	1250	0	48	Ne
42.5	Glass	1100	0	96	Ne
42.5	Glass	1050	0	96	Ne + ? Ks
42.5	Glass	1000	0	144	Ne + Ks
42.5	cryst. 1200°C to Ne	1000	0	436	$Ne(Ne_{42})$
42.5	Glass	1000	500	4.5	$Ne(Ne_{40.5})$ + weak Ks
42.5	Glass	950	0	144	Ne + Ks
42.5	Glass	900	0	700	poorly crystalline
45	Glass	1000	1000	0.5	$Ne(Ne_{45})$ + little Ks
45	Glass	900	1000	0.5	$Ne(Ne_{36.5})$ + $Ks(Ne_{18})$
45	Glass	900	500 fast quench	413	$Ne(Ne_{54})$ + Ks
45	See products of previous run	895	0	2(Hg quench)	$Ne(Ne_{45})$ + little Ks
45	cryst. 1250°C	800	0	400	$Ne(Ne_{31.5})$ + $Ks(Ne_{12})$
45	Glass	600	1000	64	$Ne(Ne_{37})$ + $Ks(Ne_9)$
45	Glass	505	1000	168	$Ne(Ne_{37})$ + $Ks(ne_4)$
45	Glass	400	1000	16	Ne + Ks (?) + ?
45	Glass	400	500	500	$Ne(Ne_{71.5})$ + $Ks(Ne_2)$
47.5	Glass	1250	0	48	Ne
47.5	Glass	1100	0	96	Ne
47.5	Glass	1050	0	96	Ne
47.5	Glass	1000	0	144	Ne
47.5	Glass	950	0	144	Ne + Ks ?
47.5	Glass	900	0	700	poorly crystalline
52.5	Glass	1250	0	48	Ne
52.5	Glass	1100	0	96	Ne
52.5	Glass	1050	0	96	Ne
52.5	Glass	1000	0	144	Ne
52.5	Glass	950	0	144	Ne

TABLE 1 (Continued)

Initial Material		Heating conditions			Results
Composition Ne wt.%	Condition	Temp. °C	P_{H_2O} Kg/cm²	Time hours	
52.5	Glass	900	0	700	poorly crystalline
52.5	Glass	800	1000	1.5	$Ne(Ne_{30})$ + $Ks(Ne_{11})$
57.5	Glass	1250	0	48	Ne
57.5	Glass	1100	0	96	Ne
57.5	Glass	1000	0	144	Ne
62.5	Glass	1250	0	48	Ne
62.5	Glass	1100	0	96	Ne
62.5	Glass	1000	0	144	Ne
62.5	Glass	700	1000	1.5	$Ne(Ne_{30.5})$
62.5	Glass	600	500	114	$Ne(Ne_{30})$ + little Ks
67.5	Glass	1250	0	48	Ne
67.5	Glass	1100	0	96	Ne
67.5	Glass	1000	0	144	Ne
71	Glass	1250	0	48	Ne
71	Glass	1100	0	96	Ne
71	Glass	1000	0	144	Ne
71	Glass	800	1000	1.5	$Ne(Ne_{71})$
100	Glass	800	500	3	Ne
37.5	Crystalline	1520	0	2	Ne + H_4 + glass
37.5	Crystalline	1520	0	6	H_4 + trace Ne + glass
37.5	Crystalline	1500	0	2	$Ne(Ne_{30})$ + glass
42.5	Glass	1500	0	3	$Ne(Ne_{30.5})$ + glass
42.5	Crystalline	1500	0	3	$Ne(Ne_{30.5})$ + glass
47.5	Crystalline	1470	0	4	$Ne(Ne_{30})$ + glass
47.5	Crystalline	1470	0	4	$Ne(Ne_{30})$ + glass
47.5	Glass	1470	0	6	$Ne(Ne_{30.5})$ + glass
47.5	Crystalline	1470	0	6	$Ne(Ne_{30})$ + glass
47.5	Crystalline	1450	0	7	$Ne(Ne_{30})$ + glass
47.5	Glass	1450	0	7	$Ne(Ne_{34.5})$ + glass
47.5	Crystalline	1450	0	7	$Ne(Ne_{30})$ + glass
52.5	Glass	1450	0	20	Ne(Ne?) + glass
52.5	Crystalline	1450	0	20	Ne(Ne?) + glass
52.5	Crystalline	1450	0	7	$Ne(Ne_{42})$ + glass
57.5	Glass	1450	0	24	$Ne(Ne_{43})$ + glass
57.5	Crystalline	1450	0	24	$Ne(Ne_{41})$ + glass
57.5	Crystalline	1425	0	7	$Ne(Ne_{35})$ + glass
57.5	Crystalline	1425	0	6	$Ne(Ne_{36})$
57.5	Glass	1425	0	6	$Ne(Ne_{37})$
62.5	Crystalline	1425	0	7	$Ne(Ne_{40})$ + glass

* Bowen's crystalline preparation, initial condition, and length of heating unknown.

A discontinuity in the nepheline-kalsilite solid solutions, resulting in a hiatus at the top of the solvus, is postulated for several reasons: 1) The structural change is rather drastic for a continuous solid solution series since the *a*-axis of nepheline is $\sim$ 10Å whereas the *a*-axis of kalsilite is $\sim$ 5.2Å, 2) Kalsilite and nepheline belong to different space groups, 3) Quenching experiments gave a mixture of nepheline and kalsilite in the Ne_{30} mixture at 1200°C, and 4) X-ray powder patterns made at temperatures above the solvus

indicate that the change from the nepheline to the kalsilite structure takes place between Ne_{30} and Ne_{20}.

In figure 2 the solvus is shown as having a maximum temperature of 1070°C at a composition near Ne_{30}. If this is the correct relationship, 1070°C is a minimum temperature at which Ne_{30} can exist as a stable phase. Any other composition between Ks and Ne_{73} will have a different minimum temperature of stable existence as given by the solvus, and in nature, if cooling is slow and equilibrium prevails, it is anticipated that an intergrowth of kalsilite and nepheline will form at this temperature on cooling.

The rate of unmixing is such that homogeneous single phases can be obtained in many compositions by dry quenching from 1200-1400°C, but reheating these same charges at 500°C produces unmixing within a few minutes. Presumably the unmixing under these circumstances is on a submicroscopic scale and the intergrowths are similar in many respects to cryptoperthites which are well known in the alkali feldspars. Such intergrowths in feldspars can be homogenized by heating above the solvus, as can those produced in nepheline solid solutions. Unmixing and homogenization in the nephelines is so rapid that the process can be studied on the x-ray heating camera. For example, a solid solution having the composition $Ne_{37.5}$ will remain homogeneous on heating in the x-ray camera to 450-500°C at which temperature lines characteristic of kalsilite suddenly appear and the nepheline reflections diminish in intensity. At the same time the nepheline lines shift in accordance with the compositional variations caused by the separation of kalsilite. The composition of the nepheline ($\sim Ne_{50}$) and kalsilite ($\sim Ne_{15}$) formed by initial unmixing at 500°C did not reach the equilibrium values (Ne_{69} and Ne_4, respectively) in the time available for the experiments. At higher temperatures, however, the unmixed phases more nearly approached equilibrium values.

The rapid and reversible nature of initial unmixing suggested that the high temperature portion of the solvus could be located by the heating x-ray diffractometer. The following method for locating the solvus was devised and found to give reproducible results: A motor-driven variac was used to heat the specimens at a uniform rate of 150°C per hour while the spectrometer goniometer oscillated about the $(20\bar{2}2)$ reflection of nepheline and the $(10\bar{1}2)$ reflection of kalsilite. (Preliminary heating at 650°C for two hours converted the homogeneous solid solution into a mixture of nepheline and kalsilite.) An x-ray record was obtained at approximately 10-minute intervals and the temperature was recorded at 40°C intervals. As the solvus was approached, the Ne and Ks reflections merged but the resulting double reflection was broad and it was possible to locate the temperature of a point on the solvus by measuring the width of the peak. Heating above this temperature produced no further decrease in width of the reflection. The reaction was somewhat more sluggish on cooling and, as a consequence, it was difficult to detect the first broadening of the reflection. Results on compositions near the top of the solvus were comparable with those obtained by the quenching method. Satisfactory measurements were made on Ne_{20}, Ne_{30}, $Ne_{37.5}$ and $Ne_{42.5}$, and they are summarized in table 2.

The Modifications of $KAlSiO_4$.—Two types of natural kaliophilite have been described: 1) "normal" kaliophilite, and 2) "abnormal" kaliophilite. The first type was described by Bannister and Hey (1931), and the second was reported by Tilley and Henry (1953) and described by Dr. A. M. B. Douglas (personal communication). The abnormal kaliophilite, according to Dr. Douglas, has diffuse reflections for $k = 3n$, whereas normal kaliophilite has sharp $k = 3n$ reflections. Both types are found only in ejected blocks in volcanic rocks.

Synthetic material identical to natural kaliophilite has not been produced but a similar phase has been found in Ne_{10} mixtures heated under hydrothermal conditions (1000-1100°C at 500 Kg/cm² water vapor pressure), and also in one mixture prepared dry by Bowen (1917) at 1075°C. The synthetic material has the strong reflections of natural kaliophilite but its weak reflections are lacking. Smith and Tuttle (1957) suggested that this synthetic material may be a disordered form of the natural kaliophilite.

In 1942 Holmes discovered a new form of $KAlSiO_4$, which he named "kalsilite", in lavas from Uganda. He pointed out that the optical properties of this material are very similar to those of nepheline, and that it is probable that kalsilite has been mistaken for nepheline in potash-rich rocks. Kalsilite has been prepared synthetically and has been found in blast furnace linings (Rigby and Richardson, 1947). In the present study it was synthesized hydrothermally at temperatures below 840°C from mixes of potassium disilicate and alumina and from the high temperature orthorhombic polymorph.

TABLE 2

Heating X-ray Diffractometer Measurements on the Ne-Ks Solvus

Composition Wt. Per Cent	Initial Material	*Temperatures at Which One Phase Disappeared on Heating and Appeared on Cooling		Comments
		Heating	Cooling	
42.5	quenched Ne_{ss}	1040 ± 20	1050 ± 20	Good records.
37.5	quenched Ne_{ss}	1070 ± 20	.....	Good records; incomplete cooling data shows that reaction is reversible.
30.0	Ne_{ss} ± Ks_{ss}	1080 ± 20	.....	Good heating record; cooling data unsatisfactory showing sluggish unmixing.
20.0	Ks_{ss} ± H_{ss}	980 ± 30	.....	Fair heating record; cooling data unsatisfactory, growth of H_s occurred (probably metastable as it transformed to Ne_{ss} at lower temperature).

* The thermocouple of the heating x-ray diffractometer was calibrated by observing the known melting point of sodium metasilicate at 1089°C in separate experiments. The calibration appeared to be reproducible to ± 10°C.

The orthorhombic form of $KAlSiO_4$ was produced by Bowen (1917) who reported that it was the high temperature polymorph of kaliophilite. Bowen heated kaliophilite at various temperatures and noted that it inverted to the orthorhombic form above 1550°C. X-ray studies of Bowen's mixtures

have shown that one specimen of his hexagonal phase is kalsilite and the other is synthetic kaliophilite. This change from kalsilite to orthorhombic $KAlSiO_4$ is readily produced under hydrothermal conditions. It is reversible and takes place at about 840°C, above which it will change to orthorhombic (O_1) $KAlSiO_4$ within a few hours, and below which the O_1 form will change to kalsilite.

Presumably the inversion of kalsilite to O_1 is raised by solid solution of $NaAlSiO_4$ since kalsilite was obtained in many runs above 840°C in mixtures containing 10 percent $NaAlSiO_4$. The inversion is shown to be raised by solid solution in figure 2.

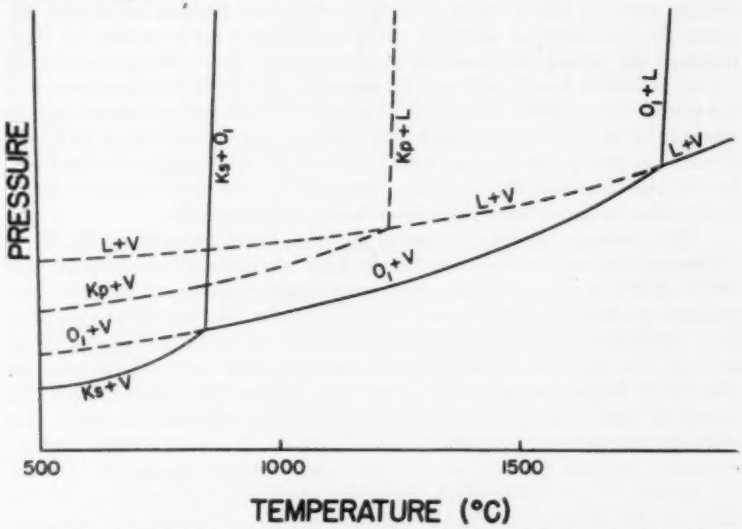

Fig. 3. Schematic pressure-temperature diagram for $KAlSiO_4$ showing the probable interrelations of the various polymorphic modifications. Dashed lines represent metastable equilibria. No attempt has been made to assign the proper slope to the nearly vertical curves. L = liquid, V = vapor, Ks = kalsilite, O_1 = orthorhombic $KAlSiO_4$, kp = kaliophilite.

Figure 3 is a schematic PT diagram showing the possible relationships between the various polymorphs of $KAlSiO_4$. Dotted lines indicate metastable equilibria.

The Modifications of $NaAlSiO_4$.—Bowen (1912) demonstrated that nepheline changed reversibly to a high temperature form, carnegieite, at 1248°C. Later studies on this form (Bowen and Greig, 1925) using differential thermal methods showed that several carnegieite specimens have a high-low inversion at 690°C, while one preparation gave two inversions on heating —226.5°C and 658.3°C, respectively. In the temperature range 1248°C to the melting temperature (1525°C) carnegieite has isometric symmetry (Bowen and Greig, 1925); below the metastable displacive transitions the symmetry is unknown, possibly triclinic.

A plot of lattice parameters versus composition for the nepheline solid solution series (see Smith and Tuttle, 1957, fig. 3) is a straight line for compositions from $Ne_{7.0}$ to Ne_{90}, but extrapolation to Ne_{100} does not intersect the values for the lattice constants of Ne_{100}. The difference between the lattice parameters obtained by extrapolation and those measured on Ne_{100} cannot be regarded as due to experimental error, although the difference is not large. Also the lattice dimensions of Ne_{100} prepared by hydrothermal crystallization of glass differ from those of Ne_{100} crystallized dry. Further differences between Ne_{100} prepared hydrothermally and dry were noted in a differential thermal study; material prepared dry showed a reversible and reproducible thermal effect at 850°C whereas no heat effect was detected in samples prepared by hydrothermal methods. High temperature x-ray studies of Ne_{100} prepared dry showed an inversion at about 900°C, at which temperature the (22.0) reflection broadened, and the intensity of the (30.0) diminished considerably. Above 900°C the (22.0) line split into two resolved peaks and the intensity of the (30.0) reflection fell to approximately 10 percent of its original value. A similar change was noted in $NaAlSiO_4$ prepared hydrothermally, but in this material the inversion temperature was approximately 100°C higher. The inversion takes place reversibly in both materials.

The inversion of Ne_{100} prepared dry was found to occur at 850°C by differential thermal analysis whereas the high temperature x-ray method gave 900°C. 850°C is the preferred value because of uncertainties in the exact temperature at which the (22.0) peak began to broaden, and because the temperature distribution throughout the sample in the x-ray heating camera is not uniform. The inversion in the hydrothermally prepared material is presumably about 50°C higher as indicated by the x-ray studies. The inversion is of the displacive type (Buerger, 1947) as shown by the reproducible rapid heat effect in the material crystallized dry. The lack of a heat effect in hydrothermally prepared material may be the result of slight changes in composition during crystallization which render the inversion somewhat sluggish. The effect of solid solution of albite (resulting from loss of alkalies, for example) on the inversion was not investigated.

Complete x-ray patterns were taken above and below the inversion. Those taken below had hexagonal symmetry, and those taken above could be indexed by assuming orthorhombic symmetry.

The effect of solid solution on the inversion temperature could not be determined since no evidence of the symmetry change was observed in the adjacent Ne_{90} mixture on heating to 1230°C. Solid solution must have a drastic effect on the inversion but it is not known whether the inversion temperature is raised or lowered. In general, it is expected that hexagonal nepheline ($Na_3KAl_4Si_4O_{16}$) would be more soluble in the low temperature hexagonal form than in the high temperature form, thereby raising the inversion. A not impossible tie-in with the nepheline-carnegieite field boundary is shown in figure 2.

The variability of x-ray properties of $NaAlSiO_4$ (Ne_{100}) prepared by dry and hydrothermal crystallization has a counterpart in the soda feldspars studied by MacKenzie (1952). He found that high temperature albite becomes

monoclinic on heating to about 920°C whereas hydrothermally prepared material did not become monoclinic on heating to the melting temperature (1118°C). The probable relations between the polymorphs of $NaAlSiO_4$ are shown schematically in Figure 4.

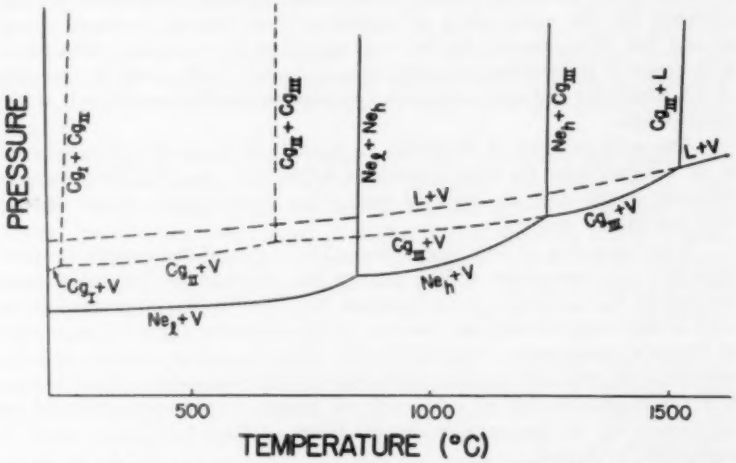

Fig. 4. Schematic pressure-temperature diagram for $NaAlSiO_4$ showing the probable stable and metastable relations for the various polymorphic forms. L = liquid, V = vapor, Cg = carnegieite, Ne = nepheline. Carnegieite may have two sharp inversions corresponding to those shown (226.5 and 658.3°C), or it may have a complex differential thermal heat effect at 690°C which may or may not represent a single inversion (see Bowen and Greig, 1925). It is probable, therefore, that carnegieite may have metastable forms not shown.

The Compound $Na_3KAl_4Si_4O_{16}$.—Buerger, Klein, and Hamburger (1947) found that the crystal structure of nepheline is based on a distorted tridymite type framework where half the Si atoms are replaced by Al and the charge is balanced by the entrance of Na and K atoms into the voids. Four different voids exist; one is large and especially suitable for K atoms, and the other three are small and suitable for Na atoms. Buerger (1954, p. 607) concluded that the natural formula of nepheline is $Na_3KAl_4Si_4O_{16}$, rather than $NaAlSiO_4$.

Evidence that $Na_3KAl_4Si_4O_{16}$ is a compound, and not a single composition in a solid solution series, is offered by the x-ray studies (Smith and Tuttle, 1957, table 2 and fig. 3) which demonstrate that the lattice parameters of nepheline solid solutions show a break at $Na_3KAl_4Si_4O_{16}$ when plotted against composition. *Such a break is unknown, and perhaps impossible, in a continuous solid solution series in which no phase transformations are involved.*

The $Na_3KAl_4Si_4O_{16}$ composition is therefore believed to be unique in the solid solution series between $NaAlSiO_4$ and $KAlSiO_4$. The uniqueness arises from the structural relations for this composition where the K and Na atoms are arranged in different sites in the structure and when the composition is

changed by substituting K for Na or Na for K, the uniqueness is lost as some K atoms must occupy sites formerly occupied only by Na atoms or some Na atoms must occupy sites formerly populated only by K atoms. This character of the $Na_3KAl_4Si_4O_{16}$ composition manifests itself in at least three ways: 1) A break in the lattice parameters when plotted against composition, 2) The tendency for the composition of nepheline from plutonic rocks to group around this value, despite the fact that nepheline from volcanic rocks has a wide range in composition on either side of $Na_3KAl_4Si_4O_{16}$, and 3) One side of the miscibility gap approaches this composition but does not cross it at low temperatures.

The solid solution of $NaAlSiO_4$ in $Na_3KAl_4Si_4O_{16}$ is therefore believed to be different from the solid solution of $KAlSiO_4$ in $Na_3KAl_4Si_4O_{16}$ because different atomic sites are involved and at low temperatures we are dealing with two binary systems.

It is interesting to note that Bowen (1917) located the eutectic between nepheline and carnegieite at 72.5 percent Ne, and that the theoretical composition of $Na_3KAl_4Si_4O_{16}$ is 72.9 percent Ne. This may be fortuitous but, in view of the compositional uncertainties in these materials and the short time of Bowen's experiments (one-half hour), it is conceivable that the eutectic represents the congruent melting temprature of the compound $Na_3KAl_4Si_4O_{16}$. It is not impossible that the compositional changes in the mixtures could be responsible for the incongruent decomposition of $Na_3KAl_4Si_4O_{16}$ at 1380°C as reported by Bowen.

The New Phase, Tetrakalsilite (H_4).—A provisional composition for this phase is $K_3NaAl_4Si_4O_{16}$ (23 wt. percent Ne), and it is likely that the composition varies on either side of this as shown in figure 2.

Conflicting evidence has been obtained on the stability of tetrakalsilite; quenching experiments on the Ne_{20} and Ne_{30} mixtures indicate a stability field as shown in figure 2, while x-ray heating runs on the Ne_{20} suggest that it may be a metastable phase formed during the quench. The fact that crystals large enough for single crystal studies were grown near the solidus and that kalsilite was converted to tetrakalsilite in long runs at high temperature (table 1) suggest that it is a stable substance. On the other hand, kalsilite having the composition Ne_{20} was observed on the x-ray heating camera to change to tetrakalsilite on cooling below 1000°C and at still lower temperatures the tetrakalsilite changed to nepheline and kalsilite. These latter observations suggest that tetrakalsilite may form metastably at low temperatures. It has not been possible to make x-ray heating camera experiments in the temperature range 1200-1400°C to resolve this dilemma.

DISCUSSION

Petrologically, the two most significant aspects of the $NaAlSiO_4$—$KAlSiO_4$ system are the solvus and the possibility that $Na_3KAl_4Si_4O_{16}$ is a compound at subsolidus temperatures. It is interesting that only a short time after the solvus was discovered (Smith and Tuttle, 1952) Sahama (1953) described intergrowths of nepheline and kalsilite in volcanic rocks from North Kivu, Belgian Congo, which he regarded as having formed by unmixing. The

intergrowths occur in phenocrysts and are complicated by having a core of "perthitic" kalsilite containing lamellae of nepheline in parallel alignment surrounded by homogeneous nepheline. The boundary between the "perthitic" core and the nepheline rim is very irregular. The nepheline lamellae in the intergrowths have a composition of Ne_{73}, and the kalsilite component carries approximately 3 percent Ne. At 400°C kalsilite and nepheline having the compositions Ne_2 and Ne_{72}, respectively (fig. 2) are in equilibrium. Thus, the compositions of the naturally co-existing kalsilite and nepheline are precisely those to be expected at low temperatures from the experimentally located solvus.

The nepheline rims of the phenocrysts have essentially the same composition as the nepheline lamellae in the "perthitic" core (i.e., Ne_{34}), and Sahama (1953) suggested that the core crystallized early and unmixed while or after the rim was precipitated. It is not readily apparent how a magma could precipitate a nepheline carrying 34 percent Ne, and then suddenly precipitate nepheline having a composition of Ne_{72} with an irregular boundary between the core and rim. It is more likely that the *rim* owes its origin to unmixing or some reaction process.

Miyashiro (1951) pointed out that nephelines in plutonic and extrusive rocks show distinct compositional differences. Nephelines in volcanic rocks have a wide range in kalsilite content, whereas nephelines from plutonic rocks are more restricted in composition. He attributes the difference to a structural control of composition at low temperature resulting from the ordering of K and Na atoms. Tilley (1952, 1954, 1956) reported similar findings and tied the nepheline composition to the normative Q-Ne-Kp in the host rock, thus giving a more complete picture of chemical changes in the composition of the nepheline as a function of host rock composition. Tilley further showed that the nepheline of an extrusive rock, on cooling, or by subsequent metamorphism, must change composition. In the specific example chosen (Tilley, 1954, fig. 3, p. 73) the high temperature nepheline solid solution was rich in soda and it was suggested by Tilley that potash would be added to the nepheline by reaction with feldspar, thus the final composition of the nepheline would tend to approach the composition $Ne_3KAl_4Si_4O_{16}$.

The relations reported by Tilley together with the stability relations in the $NaAlSiO_4$—$KAlSiO_4$ and $NaAl_2Si_2O_8$—$KAlSi_2O_8$ systems permit the drawing of a series of schematic isothermal, isobaric diagrams depicting phase relations at several temperatures. Four such schematic diagrams are shown in figure 5. These diagrams are intended to illustrate equilibrium relations below the solidus, although a small area of figure 5A, which is at a temperature just above the binary nepheline-kalsilite solvus, is in fact above the liquidus.

The range in composition of all analyzed nephelines suggests that ternary solid solution is extensive at high temperatures and an isotherm near the top of the nepheline-kalsilite solvus must have the general relations shown in figure 5A. Solid solution of albite in $NaAlSiO_4$ is extensive as shown by Greig and Barth (1938), and the region of ternary solid solution is shown as beginning at the maximum solid solution reported by them. It is probable that

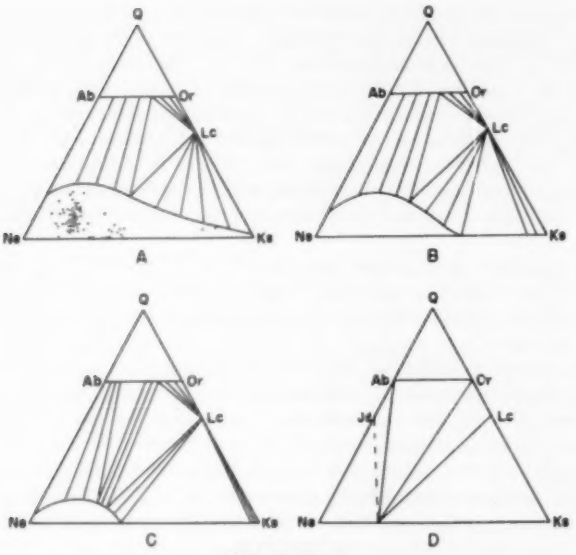

Fig. 5. Schematic isothermal, isobaric, diagrams for the nepheline-kalsilite-quartz system showing the probable phase relations at different temperatures.

(A) Temperature above the Ne-Ks solvus. At this high temperature the nepheline structure takes large amounts of silica into solid solution, the extent of which is shown by the dots representing analyzed nephelines and kalsilites. This extensive solid solution is undoubtedly responsible for the many attempts to assign a "formula" to nepheline.

(B) Temperature below the top of the nepheline-kalsilite solvus but above the albite-orthoclase miscibility gap. At this temperature the extent of solid solution is less and leucite can co-exist with nepheline and kalsilite.

(C) Temperature below the top of the feldspar miscibility gap. Two alkali feldspars can now co-exist with nepheline.

(D) Section at low temperatures in which a miscibility gap between nepheline ($Na_3KAl_4Si_4O_{16}$) and the end member $NaAlSiO_4$ is postulated. Jadeite (Jd) will appear at high pressures and if nepheline is stable at this presure, the join jadeite-nepheline will be present.

the maximum silica content will occur in the binary system $NaAlSiO_4$—$NaAlSi_3O_8$ at these high temperatures although it is possible that a maximum will be found along the join $Na_3KAl_4Si_4O_{16}$—albite, as shown in figure 5A. The magnitude of solid solution of nepheline in alkali feldspars is unknown but, in view of the results of Greig and Barth (1938, p. 106-107), it is probably not more than a few per cent; consequently no solid solution of this type is shown in figure 5A. The extent of solid solution between potash feldspar and leucite is also unknown but if solid solution were extensive, it would have been detected in analyzed sanidine and co-existing leucite.

Figure 5A is characterized by a large area of nepheline solid solution, a three-phase region in which potash feldspar solid solution, leucite, and

nepheline solid solution co-exist, and three two-phase regions—one in which feldspar solid solution is in equilibrium with nepheline solid solution, another in which feldspar solid solution and leucite are in equilibrium, and a third in which leucite is in equilibrium with nepheline-kalsilite solid solution.

Figure 5B shows the relationship expected at a temperature below the top of the nepheline-kalsilite solvus and above the albite-potash feldspar solvus. At this temperature a second area in which three solid phases co-exist appears; the three phases are leucite, kalsilite solid solution, and nepheline solid solution. The field of nepheline solid solution is shown to be somewhat smaller than at the higher temperature of the previous isotherm.

At temperatures below the top of the alkali feldspar miscibility gap (fig. 5C) a third region in which three solid phases co-exist appears; the phases are albite solid solution, potash feldspar solid solution, and nepheline solid solution. At these low temperatures, corresponding perhaps, to temperatures of final crystallization of nepheline syenites, the area of nepheline solid solution is considerably reduced as indicated by the analyses of nepheline from rocks carrying albite and microcline. The phase relations in the area $NaAlSiO_4$—albite—$Na_3KAl_4Si_4O_{16}$ cannot be deduced from the plutonic rocks because rocks having the appropriate composition have not been studied. An alternative diagram in which $Na_3KAl_4Si_4O_{16}$ is shown as a compound with a solvus between it and $NaAlSiO_4$ is illustrated in figure 5D. It is not possible, with the data on hand, to decide which of these two diagrams (figs. 5C or 5D) represents the closest approach to the phase relations at low temperatures.

Perhaps the most significant feature of this low temperature isotherm is the fact that a join from albite to $Na_3KAl_4Si_4O_{16}$ may be present in view of the contrasting composition of nepheline from extrusive and plutonic rocks (Tilley, 1954). This join separates the stability field of jadeite from the area of the diagram in which the bulk composition of the plutonic alkaline rocks falls, and although it is commonly assumed that the extreme rarity of jadeite is a result of its high pressure stability, this join precludes the stable occurrence of jadeite in these rocks as long as albite and nepheline are stable. If nepheline is stable at the pressure at which jadeite becomes a stable phase in this system, figure 5D will be modified by the appearance of jadeite and the join jadeite—$Na_3KAl_4Si_4O_{16}$. Such a join gives rise to two three-phase assemblages: albite—jadeite—$Na_3KAl_4O_{16}$ and jadeite—$Na_3KAl_4Si_4O_{16}$—$NaAlSiO_4$. This relationship permits the stable co-existence of jadeite with nepheline and albite. Some light could perhaps be thrown on the nepheline problem by studying the composition of nephelines associated with jadeite.

A series of unmixing reactions in nepheline are probable in view of the wide range of compositions of nepheline solid solutions in high temperature extrusive rocks. The following phases may unmix from a nepheline solid solution:

nepheline solid solution → nepheline + kalsilite
→ nepheline + kalsilite + leucite
→ nepheline + leucite
→ nepheline + leucite + potash feldspar

→ nepheline + potash feldspar
→ nepheline + potash feldspar + albite
→ nepheline + albite

The final composition of the nepheline will approach the composition $Na_3KAl_4Si_4O_{16}$. The amount of unmixing in a given example will depend upon the extent of solid solution at high temperature and on the opportunity for obtaining equilibrium during cooling. The unmixed phases may migrate from the nepheline host crystals, particularly if volatile materials are present to flux the reaction, or they may unmix within the nepheline giving a "perthitic" type structure.

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PROPERTIES OF WATER. PART III. SPECIFIC VOLUME OF LIQUID WATER TO 100 C AND 1400 BARS*

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ABSTRACT. Published values for the specific volume of liquid water at modest to high pressures range widely. New measurements of the specific volume of water to 100 C and 1400 Bars are presented with the hope of resolving the conflict. Surprisingly, the present data agree best with some of the earliest published work, that of Amagat, and is most in disagreement with some of the more recently published values.

INTRODUCTION

The compressibility of liquid water has been measured by several workers over the last 70 years. The list of investigators includes almost the entire roster of people who have done extensive work in the field of high pressures. Data are available from Amagat, Ekman, Bridgman, Tammann and Jellinghaus, Adams, Gibson and Loeffler, and Smith and Keyes.

The disagreement among these various investigators is large, compared with the apparent precision of the experiments, as is immediately apparent from an inspection of the tables published by Birch (1942). The present investigation was undertaken with the hope of resolving this conflict.

DESCRIPTIONS OF APPARATUS

The apparatus used in the present investigation has been described in detail elsewhere (Kennedy, 1950, 1954). The bomb, of conventional design and seal, was made of Inconel-X alloy. Its volume was approximately 170 cc, as measured by filling with water under pressure and weighing; precision of the method was about 0.05 percent. In operation the bomb was immersed in a stirred oil bath and kept at a desired temperature by a precision mercury regulator. A hand compressor, similar in principle to the one described by Keyes (1933), copied from one designed by Benedict (1939), was used to inject water into the bomb. Pressures were measured on a Heise Bourdon tube gauge with a range of 1400 bars. This gauge was calibrated with the deadweight piston gauge used in the Massachusetts Institute of Technology steam program (Keyes 1933, p. 529), and kindly loaned us by Keyes. The piston diameter of the dead-weight gauge was measured by interferometer, and the clearance of the piston was measured by the leakage rate of pure carbon tetrachloride at a known temperature and pressure according to the technique of Meyers and Jessup (1931). The effective area of the piston was calculated by the methods described by Meyers and Jessup.

EXPERIMENTAL METHODS

The present investigations did not attempt an absolute determination of the specific volume of liquid water at any pressure or temperature. The data of Tilton and Taylor (1937) and Owen, White and Smith (1956), on the specific volume of water at one bar pressure in the temperature range 0 to 100 C were accepted, and only relative changes of specific volume as a function

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of pressure were determined. In practice, the bomb was placed in an oil bath thermostated at some temperature between 0 C and 100 C, and filled initially to a measured low pressure. Then the quantity of water required to change the pressure from one measured value to another was determined by means of the calibrated screw of the compressor. The specific volume of water in the bomb was then calculated using this quantity, the volume of the bomb, its thermal coefficient of expansion, and its pressure dilation. The increments of water were summed over all lower pressure intervals to obtain the specific volume at each pressure. This measurement was very precise: for a given temperature and pressure it was reproducible with a relative standard deviation of about 1 part in 5,000, which corresponds to a variation of at most only 0.00001 in the specific volume.

The isothermal runs were spaced at intervals of 10 C. The temperature of the oil bath was measured with a mercury-in-glass thermometer. It was not necessary to know the temperature of the bomb with extreme accuracy, but only that during a series of runs the temperature of the bomb remained constant within very narrow limits. The thermometer indicated that the thermostat varied no more than 0.02 C during a run, although absolute temperature was known to only about 0.1 C. However, the compressibility does not vary appreciably with a temperature change of 0.1 C, so no significant error is introduced by this small uncertainty in the temperature of the bomb.

Several factors have about equal effect in limiting the accuracy and precision of our determinations of specific volume: determination of bomb volume, calibration of the Heise gauge, and determination of the weight of water added. Known errors in these factors affect the final specific volume by 0.0001 to 0.0002.

The measured specific volumes were tabulated and smoothed. The final smoothing involved changes of a maximum of 0.0002 cc/g in specific volume, and of only 0.0001 cc/g over most of the tables.

RESULTS

The specific volumes of water in cc/g at temperatures ranging from 0° to 100° and at pressures ranging from 1 bar to 1400 bars, are presented in table 1. These tables are smooth in both the first and second differences and seem thoroughly consistent internally.

The most striking observation concerning the volume data is the effect of pressure on the thermal expansion. The quantity $\Delta^2 v/\Delta T \Delta p$ is positive at temperatures below 40 C, zero near 40 C or 50 C, and negative at higher temperatures. These observations confirm those of Amagat (1893, p. 553).

COMPARISON OF RESULTS WITH OTHER INVESTIGATORS

The results of this investigation are compared with the tabulated results of Amagat (1893, p. 544, as corrected by Dorsey (1940, p. 207) to an absolute specific volume of 1.0000 ml/g at 4 C and 1 atm) and Smith and Keyes (1939, table V) in figures 1 and 2. Deviations between the data of Amagat, Smith and Keyes and our data are shown in parts per 10,000 of the determined specific volume as a function of pressure. The compressibility is known

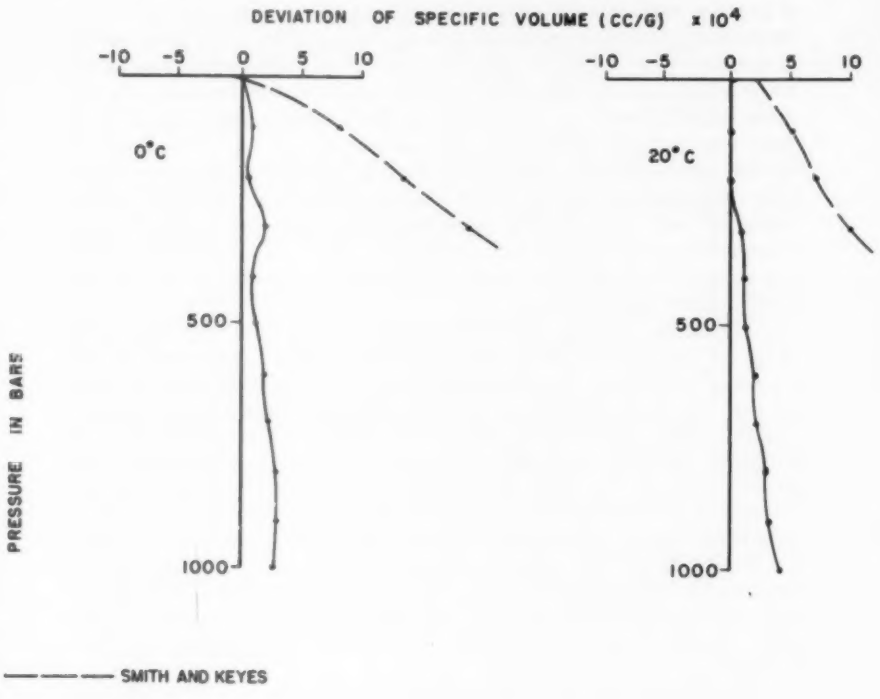

Fig. 1. Comparison of data on specific volume of water (values of other observers less values of this investigation in cc/g x 10^4).

accurately enough to interpolate their readings in atmospheres for comparison with our readings in bars. At 0, 20, 60, and 100 C, where the comparisons have been made, the Amagat data everywhere agree with the data herein presented to better than 1 part in 2,500. At 1000 bars the specific volumes of Amagat are consistently higher than ours by about that amount. The agreement is particularly close, in view of the fact that our data have only been smoothed to 1 part in 10,000, and the adjustment on smoothing has been as large as 1 part in 5,000. All the data are in substantial agreement at 60 and 100 C, although our data agree with those of Smith and Keyes somewhat better than with those of Amagat, particularly if the measured rather than the tabulated values of Smith and Keyes are used. However, the data of Smith and Keyes at 0 C and 20 C diverge sharply from both the present results and those of Amagat. Smith and Keyes made no measurements below 30 C, and the values that they tabulate in this region are extrapolations, by an empirical equation, from their measurements above 30 C. They also extrapolate down to 1 bar and tabulate values differing considerably from those already well-

known in the range 0-100 C. It is surprising that Smith and Keyes made this extrapolation without guidance from the earlier data.

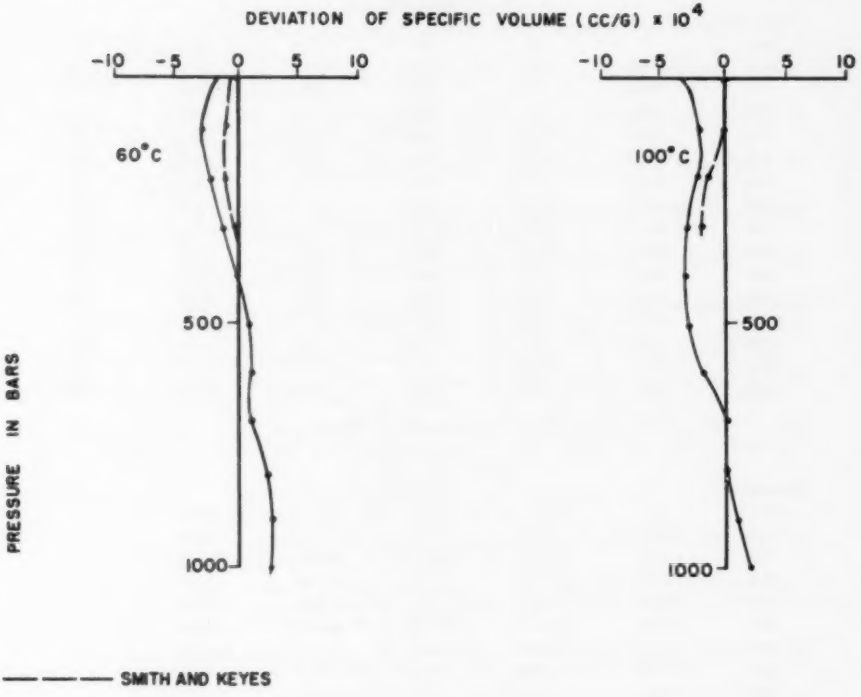

Fig. 2. Comparison of data on specific volume of water (values of other observers less value of this investigation in cc/g x 10^4).

The data of Gibson and Loeffler (1941) extend from 25 to 85 C and to 1000 bars, but they cannot be compared directly with the above data as they are tabulated at intermediate temperatures. However, interpolation of the data of Gibson and Loeffler at 20 and 80 C affords an approximate comparison. Their values lie very close to those of Amagat.

The investigations of Bridgman (1912, 1931, 1935) and of Adams (1931) were principally aimed at high pressures, and make no claim to high accuracy at the pressures here considered. The various determinations of Bridgman scatter widely but are generally further from our values than are those of Amagat, while Adams' one isotherm agrees closely with Amagat and with Gibson and Loeffler.

The precision and consistency of the various water data are discussed in further detail by Eckart in another paper of this series (Eckart, 1958).

TABLE 1

Specific Volume of Water

cc/g

Pressure bars	Temperature, deg C					
	0	10	20	30	40	50
1	1.0001	1.0003	1.0018	1.0043	1.0078	1.0121
100	.9950	.9956	.9972	.9998	1.0034	1.0077
200	.9902	.9911	.9928	.9954	.9990	1.0033
300	.9855	.9866	.9885	.9912	.9948	.9991
400	.9809	.9823	.9843	.9872	.9908	.9951
500	.9765	.9781	.9802	.9832	.9869	.9911
600	.9722	.9740	.9763	.9793	.9830	.9873
700	.9681	.9701	.9724	.9754	.9792	.9835
800	.9640	.9662	.9686	.9717	.9755	.9798
900	.9601	.9624	.9650	.9682	.9720	.9763
1000	.9563	.9587	.9614	.9647	.9686	.9729
1100	.9526	.9551	.9579	.9613	.9652	.9695
1200	.9490	.9517	.9546	.9580	.9619	.9662
1300	.9455	.9483	.9513	.9548	.9587	.9630
1400	.9421	.9450	.9481	.9516	.9555	.9598

Specific Volume of Water

cc/g

Pressure bars	Temperature, deg C				
	60	70	80	90	100
1	1.0171	1.0227	1.0290	1.0359	1.0434
100	1.0127	1.0182	1.0244	1.0311	1.0384
200	1.0083	1.0138	1.0199	1.0265	1.0336
300	1.0041	1.0095	1.0155	1.0220	1.0290
400	1.0000	1.0053	1.0112	1.0176	1.0245
500	.9960	1.0013	1.0071	1.0134	1.0201
600	.9921	.9974	1.0031	1.0093	1.0158
700	.9883	.9935	.9991	1.0052	1.0117
800	.9846	.9897	.9953	1.0013	1.0076
900	.9810	.9861	.9916	.9975	1.0037
1000	.9775	.9825	.9879	.9938	.9999
1100	.9741	.9790	.9844	.9902	.9962
1200	.9708	.9757	.9810	.9867	.9927
1300	.9676	.9724	.9776	.9833	.9892
1400	.9644	.9692	.9743	.9799	.9858

ACKNOWLEDGMENTS

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addition, Dr. Carl Eckart offered many useful suggestions during the course of the investigation.

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Note added in proof: The data of table 1 above are referred to in part II by Eckart (1958, p. 226) with an erroneous reference to part I of this series.

DISCUSSION

POSSIBLE PERMIAN CLIMATIC ZONATION AND ITS APPLICATION

GEORGE W. BAIN

The recent article on "Possible Permian climatic zonation and its applications" (Stehli, 1957) sets forth three conclusions:

1. A sharp boundary to the apparent ranges of a number of Permian marine invertebrates occurs between 50° and 60° N wherever data permit its location.
2. The faunal boundary parallels the earth's present equator and, if truly caused by temperature, precludes the possibility of changes in the position of the poles with respect to the major land masses of the northern hemisphere.
3. Also precluded is the possibility that the crust or mantle has shifted its position relative to the core.

The data submitted in support of the three conclusions should be examined carefully before the generalizations are accepted and used to develop further deductions. Each is examined in the above order.

1. The boundary is not sharp.

The distribution of selected foraminiferal groups are repeated in figure 1 and boundary lines are drawn to include within a zone all occurrences except that in New Zealand which is unique in other ways than its isolation. This zone is interrupted through the middle of the Phosphoria Sea in America, the Western Uralian Sea in Europe and across the Sikkim Gateway in India. The gaps detract from both sharpness of border and continuity of

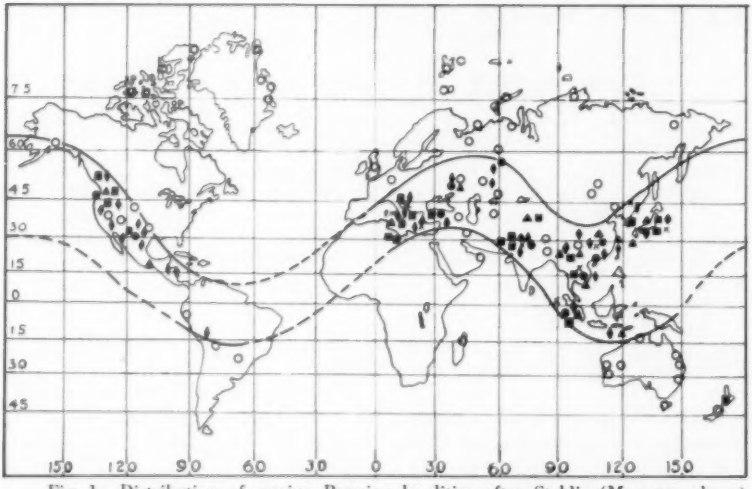

Fig. 1. Distribution of marine Permian localities after Stehli. (Mercator chart.) The blank circles represent marine Permian localities without foraminifera of the selected groups. The solid symbols are foraminifera localities similar to those of figure 3.

fossil zone and are as valid for any of the brachiopod groups selected as for the foraminifera.

2. The faunal boundary is undulatory.

The faunal boundary has a complex undulatory form and two domains, extending hardly beyond the topics, alternate with two reaching to 60° N (fig. 1). The zone, in the case of both selected brachiopoda and foraminifera groups for the recorded occurrences, has only 30° to 50° width; the band is deflected southward in both the Atlantic Ocean and the east Asia area and northward in the Ural Mts. and the eastern Pacific Ocean; the meridians of inflection are approximately 175° W, 70° W, 35° E and 110° E. The data presented shows that both the north boundary and the axis of the fossil belt is not parallel to the present equator but is undulatory at about 90° ± 15 recurrence. The axis to the zone of selected fossil groups is tangent to the present equator at 70° W and 115° E and otherwise lies north of the equator. Enough noncritical Permian localities lie in areas outside this zone to indicate that the form is not chance sampling. The northward displacement of the entire pattern can be explained in three ways and the first is much to be abhorred; they are:

a. The lands originally lay farther south and the entire equator slipped northward about 23°. (This would set aside conclusion 3.)
b. Inadequate data in the southern hemisphere automatically displaces the statistical center of the zone to northward. (This seems to be a rational explanation.)
c. The southern hemisphere of today had abnormally low temperature during Permian time. (It had two areas of continental glaciation.)

The distribution form approximates a double sine curve. A great circle on a globe, if transferred to a Mercator map, becomes a single sine curve,

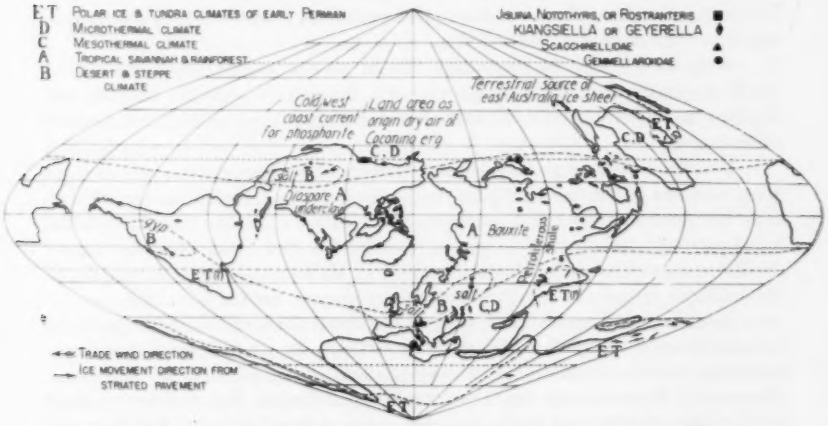

Fig. 2. Permian climatic map with equator centered on tropical rain-forest indices. (Sinusoidal chart.)

The solid symbols show occurrences of selected brachiopod groups (locations after Stehli).

and the angular amplitude of the sine curve is the inclination of the great circle to the earth's equatorial section. The existence of a double sine curve leaves explanation of the pattern, very complex if geometric at all.

The equatorward bulges in the northern hemisphere correspond to the great Permian land masses of the Canadian Shield in North America and Angaraland in Eurasia. One southern hemisphere bulge is at the great African Gonwanaland mass. Unfortunately the other is in the South Pacific region where amount of direct data is inconsequential.

3. It is very probable that the crust has shifted relative to the core.

The data given by Stehli was plotted on a transparent globe for additional study. Paleosols, salt deposits, desert dunes, glacial striae and ocean current patterns for the Permian were added to his fossil data; these have definite meaning and indicate latitude (Bain, 1953). Drying air is descending high level air at Horse Latitude and Polar high pressure areas; these areas are determined principally by heat supply and earth rotation and so remain more fixed in position than the thermal equator. The warm area between the Horse Latitudes and the equatorial low pressure zone becomes a desert with many dune areas on land and evaporite deposits on the sea margin (Bain, 1956a, p. 759; Bain, 1956b, p. 273). Wet air is principally ascending air at the equatorial and Polar Front low pressure regions; it nurtures a dense plant growth with laterite soil in the tropics and podzol in the Polar Front belt. Air movement poleward from the Horse Latitudes is easterly and equatorward therefrom is westerly; the air motion controls the principal shallow ocean currents and thereby any migration of plankton. The best equator for all these statistics was selected and the data on the globe was transferred to a sinusoidal projection (figs. 2 and 3, Bain, 1956c, p. 63). This equator passes through Andean South America, Florida, Hudson's Bay, the Arctic Archipelago, Western China, Indonesia and Antarctica. The north pole was near Samoa at 7½°S 165° W.

Angaraland and the Canadian Shield at one place and Antarctica at another split the equatorial drift, (fig. 3); the former gave the Behring and Uralian northward spread of faunas and Antarctica gave the New Zealand occurrence.

General comment and additional possibilities.—The entire topic of mapping climates of the geologic past requires much more varied sources of data than has been used by Stehli or most earlier authors (Menner) and offers the same potential for advance in geologic knowledge that came when the dogma of Werner was abandoned for the realism of Hutton and Lyell. A restudy of the multiple sources of data bearing on the climatic record is in order.

To this end, I visited and studied the Australian and African glaciated pavements and found that the striation criteria as applied need some considerable revision in instances. The great dune areas, salt deposits and phosphorites of Permian age strata in the Western United States have received critical and coordinated study and their several features show that they are North Trade Wind belt accumulations and that the Permian age parallels of latitude almost coincide with the present meridians of longitude (Bain, 1956c, p. 63).

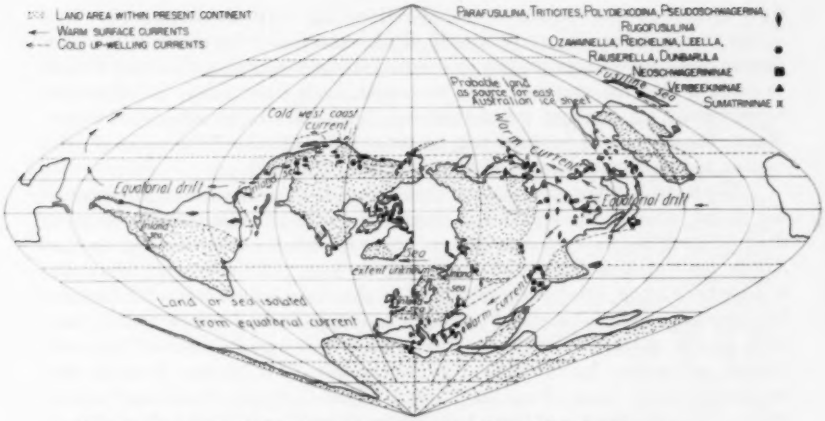

Fig. 3. Generalized Permian paleogeography. (Sinusoidal chart.)
Known warm and cold currents and the principal persistent land masses (stippled) are shown. Solid symbols indicate selected foraminifera occurrences (after Stehli).

The Permian Age equatorial drift moved from Antarctica past West Australia and impinged on Angaraland to divide just as the present Atlantic Equatorial Drift divides on South America (fig. 3). One branch passed along east Asia to northern Alaska; the other followed the Tethys Sea to central Russia and the Mediterranean. The South American part of the Equatorial Drift passed north of Antarctica and into the New Zealand area as it swung poleward towards Samoa. The full return circulation has not been mapped, mainly due to lack of data on Saharan Africa.

Late Paleozoic plankton migration path anomalies, such as from Europe to America against the present North Atlantic drift, disappear with use of this current pattern. Protozoa, and the young of all the lower animals of the Paleozoic, migrate only with the warm currents. Paucity of fossils in the pelletal part of the Phosphoria Formation is expectable in a cold upwelling current that is the return of the New Zealand warm current.

In yet another field, liberal study of past climates affords a tool to lay a "ghost" to rest or bring forth a real object. Much as one may abhor continental drift the equatorward position of glacial pavements in the southern hemisphere impressed du Toit (1937) and were one item affecting Carey's viewpoint (1955). Many reported tillites are open to question; certainly continental tills exceeding 500 feet in thickness are unknown in the Pleistocene, are not to be expected anywhere, and it is doubtful whether any conglomerate exceeding 1000 feet thickness has ice sheet origin. I have not seen the tills of India or Brazil; but descriptions of thickness and fabric, while not precluding a piedmont glacial origin, leave some doubt concerning deposition by a continental ice sheet.

A review of the entire assemblage of climatic evidence indicates recurrent polar migration, probably intermittently. Runcorn's (1955) studies of resi-

dual magnetism need studious consideration; the most discouraging feature of the results is the 50° northeasterly displacement of the position of the pole determined from European data in contrast to American data. One may be led to suspect that the magnetic pole precessed, as it does today, and that the measurement of residual magnetism was made on strata differing in age. Many climatic record features suggest progressive increase in amount of precession to the polar axis, preceding permanent polar shift.

Much as I abhor continental drift, enough evidence exists in patterns for major structures, climate and migration paths of organisms to suggest drift of earth blocks exceeding 10°, or more than twice the movement on the Alpine fault of New Zealand of the San Andreas rift of California. This drift is based not upon fitted surficial forms of the classical concept of Wegener, but involves origin for isolated bodies of incompletely understood land blocks such as the extensive Paleozoic Age continental deposits of the Falkland Islands and source for Mesozoic Age continental strata of New Zealand, the nonbasaltic rock types along the "Easter Island-Galapagos Rise" and many others. The significance of these isolated Southern Hemisphere bodies cannot be understood by "quadrangle" geology and the problems they pose in earth history are not accounted for by inaccurate restatements of traditional generalizations.

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REPLY

FRANCIS G. STEHLI

In his discussion of *Possible Permian Climatic Zonation and its Implications* Bain has raised points which require consideration. The most important of these is the alternative interpretation of Stehli's distribution plots presented in Bain's figure 1 and in pertinent comments. Bain has chosen the data for fusulinid distribution and has, by regarding this data separately and neglecting an isolated control point, been able to bound the area of occurrence with parallel double sine curves. His purpose is evidently to show that Stehli's interpreta-

tion of the data is only one of several that are possible. The writer agrees that this is the case and that the data are inadequate to permit one unquestionable solution. Interpretation of the data becomes a matter of selecting the most reasonable of alternative hypotheses.

Reference to Bain's figure 1 shows that he was able to interpret the data as he did because of the absence of control in the Atlantic, Indian and Pacific ocean basins, eastern North America and Africa. Areas lacking outcrops of Permian rocks will always lead to a certain degree of uncertainty in interpretation. However, some measures can be taken to reduce this uncertainty. First, as the information is admittedly scattered, all the data originally presented have been assembled on figure 1 of this reply in the belief that this procedure will lead to more objective results than Bain's use of the data for a single group. Secondly, in an attempt to gain more control in the southern hemisphere, the location of important finds of large terrestrial Permian reptiles has been added to figure 1. It is assumed that these animals like other reptiles were poikilothermic and controlled their body temperature behaviorally rather than physiologically. Save for a few aquatic forms which extend into the warmest part of the warm temperate zone the distribution of large reptiles is limited today to the tropics and subtropics. It is assumed that similar temperature limitations controlled the distribution of Permian reptiles. It will be noted that the assembled data together with the data for reptile distribution furnish considerably more control than was used in Bain's plot and that his interpretation can not be defended.

Let us now examine figure 1 and attempt to derive from it the most reasonable interpretation recognizing, of course, that control is still limited by the absence of Permian rocks in the Atlantic, Pacific and Indian Ocean basins

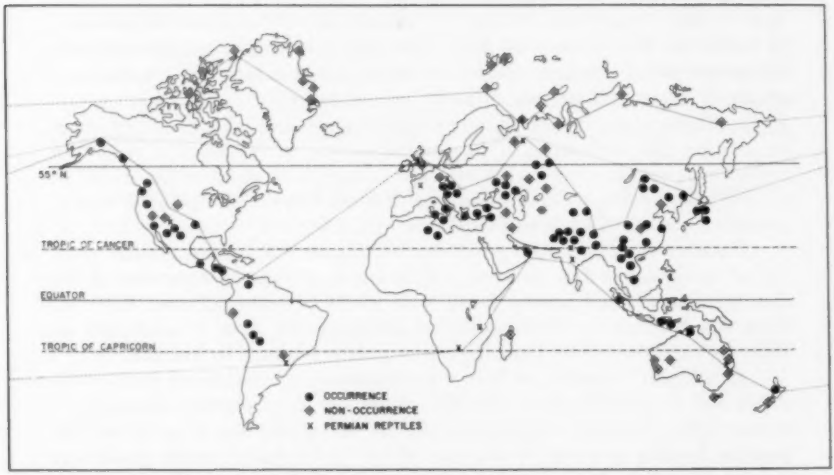

Fig. 1. World distribution of certain groups of marine invertebrates and reptiles during Permian time. (For further explanation see text.)

and in eastern North America. If we connect all the non-occurrence control points in the northern hemisphere which lie to the north of occurrence control points, we can draw a line approximating the maximum possible northward extension of the subtropical climatic zone—always assuming that the temperature significance of the organisms employed is correctly interpreted. If we now, in a similar manner, connect the northern-most control points for occurrence, we can draw a line representing the minimum possible extent of the northern margin of the subtropics. It will be noted that we have now outlined in the northern hemisphere a belt of variable width within which the northward border of the subtropics should have been located. The northern and southern margins of this belt do not parallel one another as would be expected if the subtropical margin of Permian time now traced a sinuous course on the earth's surface. Instead major deviations may be noted. They are found however only in the minimal distribution boundary and invariably coincide with areas in which there is complete lack of control. If these southward deflections due to lack of control were neglected and the four northernmost occurrence points connected by straight lines, the resulting line would very nearly parallel the maximum distribution limit. A glance at the distribution plot of modern animals known to be limited by temperature such as the reef coral plot shown by Ekman (1953, p. 5) will show that such an approach to the interpretation of areas of no control is the only reasonable one.

If attention is now given to the southern hemisphere it is at once apparent that control is still very poor. No line representing the southernmost possible limit of the Permian subtropics can be drawn as non-occurrence control points are lacking. It is possible however, to draw a line representing the minimum possible extent of the subtropical zone. There is thus delimited between this line and its northern hemisphere counterpart a definite tropical and subtropical zone. It may be seen that this belt is extremely narrow in places, for instance, in India, the Middle East and South America. Even under the earth's present subglacial climatic regime the belt of tropical and subtropical conditions is about 50 degrees of latitude in width and of grossly the same width everywhere. The local narrowness of the Permian tropical and subtropical belt seems to indicate that deflections of its boundaries are due to absence of control rather than displacement of the subtropical margin as suggested by Bain. In the Permian the tropical and subtropical climatic belt should also have been of roughly the same width throughout the world.

Possibly one could, with strike-slip faulting of sufficient magnitude, drifting of continents and/or polar migration arrive at some configuration of the continents that would satisfy the data. It would be quite different, however, from that required to satisfy published paleomagnetic data. It would also be different from that which Bain (figs. 2-3) feels is required by his data.

It is highly desirable, as Bain has commented, to consider all lines of evidence and to consider them critically. Stehli was not, however, attempting a monographic treatment of Permian climate but attempting to point out the possible bearing of a line of evidence which he felt had received insufficient consideration. The data presented represent only the information known to him at the time and may be changed by new discoveries. In the writer's opinion,

however, the paleozoogeographic data are most reasonably satisfied when interpreted as evidence of a climatic zone which, within the resolving power of the method, appears to have been parallel to the earth's present equator.

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PAN AMERICAN PETROLEUM CORP.
TULSA, OKLAHOMA

COMMUNICATION

21st INTERNATIONAL GEOLOGICAL CONGRESS

The 21st International Geological Congress will be held in the Nordic countries—Denmark, Finland, Iceland, Norway, and Sweden—in the summer of 1960. The Congress itself will be held in Copenhagen from the 15th to the 25th of August, and there will be field excursions in all the Nordic countries for two weeks before and two after the Congress.

A first circular, with response forms for those interested in attending the Congress or excursions, has been published. Copies may be obtained from the American Geological Institute, 2101 Constitution Avenue, Washington 25, D. C., or from the General Secretary, XXI Session, International Geological Congress, Mineralogisk Museum, Oster Voldgade 7, Copenhagen K, Denmark. Those requesting the circular from the AGI will be placed on a mailing list to receive travel literature and other pertinent information. Further circulars will be sent to those who respond to the first circular.

REVIEWS

A Treatise on Limnology. Vol. I. Geography, physics, and chemistry; by G. Evelyn Hutchinson. P. xiv, 1015; 228 figs., 132 tables. New York, 1957 (John Wiley & Sons, Inc., $19.50).—Limnologists have known for years that this book was in the making. The impatience of their wait was justified, for this is a great book. Apart from all other good reasons, it is great because it reflects so thoroughly its remarkable author—remarkable for combining the empirical sense of a working field naturalist with an extraordinary capacity for analysis and fantastic erudition.

The aim of this book is to give as complete an account as possible of the events characteristically occurring in lakes. This is the first sentence of Hutchinson's preface—a charmingly simple statement of the large and difficult task to which the 902 pages of the text attest, page by page. His preface continues indicatively, "The book is addressed to all who are professionally concerned with limnology, but also to biologists who may wish to know something of the physiochemical environment, mode of life, and evolutionary significance of such fresh-water organisms as they may study from quite different points of view; to geologists who are desirous of learning something of modern lakes in order that they may better interpret the record of inland waters in past times; and to oceanographers who wish to compare the results of their own science with what has been learned of the small but very individual bodies of water which make up the nonmarine part of the hydrosphere." His discussions throughout the book are fullest on subjects that "most biologists or geologists would find difficult to obtain elsewhere", and in framing these discussions he has followed the principle that "what he himself has found perplexing has probably perplexed other investigators." The full impact of this beguilingly simple statement will strike the reader as he continues. Plainly, it is not the simple things that perplex G. E. H.

A novel aspect of the book is the "undocumented, factual summary of all the main conclusions" given at the end of each chapter. These summaries are fine, compact pieces of exposition that aggregate about 47 pages. If one were to read only these—a book within a book—one would have a good idea indeed of what is known today about physical limnology.

But if one read only the summaries, one might miss the Hutchinsonian trait of covering the waterfront in literature sources. To bring the number of items in the bibliography within reasonable compass he flagged 165 items as "most convenient sources" of large amounts of earlier work. Even so the bibliography contains 1,460 titles.

Some measure of the extent to which mathematics is used is revealed by the list of 365 defined symbols, some of which are defined only by equations. Other helpful and unusual features of the book are an Index (17 pp.) of Lakes, which fixes the geographic position of each, and an Index of Organisms, which gives the order and family of each, but for some only the phylum.

The book's 17 chapters are listed here as a convenient means of showing how the subject matter is presented.

The origin of lake basins	163 pp.
The morphometry and morphology of lakes	31

The properties of water	26
The hydrological cycle and the water balance of lakes	29
The hydromechanics of lakes	116
The optical properties of lakes	60
The thermal properties of lakes	115
The inorganic ions of rain, lakes, and rivers	34
Oxygen in lake waters	78
Carbon dioxide and the hydrogen ion concentration of lake waters	38
Redox potential and the iron cycle	36
The phosphorous cycle in lakes	26
The sulfur cycle in lake waters	35
The silica cycle in lake waters	13
Minor metallic elements in lake waters	35
The nitrogen cycle in lake waters	42
Organic matter in lake waters	25

The first chapter is the longest and differs from all the others in being almost wholly descriptive. He recognizes 76 types of lakes grouped into categories such as tectonic basins, solution lakes, etc. Seepage lakes are not recognized as a numbered type, apparently because most of those known can be identified with a genetic origin. This leaves no place for the curious, small, shallow seepage lakes and ponds found sparingly in the Rocky Mountain region. These are not in glaciated areas and their origin is obscure. They have vertical margins and although only about 30 cm deep and exposed to intense insolation the water is essentially fresh and the level seems not to fluctuate. A typical example is Pickett Lake in southern Fremont County, Wyoming.

All subsequent chapters are analytical—intensely so. But this is neither new nor surprising for Hutchinson. All his work is characterized by a habit of analyzing results of investigation, his own or anyone's; and by his capacity to give the findings perspective by holding them against existing knowledge, to add something of significance, or to indicate the direction further inquiry should go. The fantastic erudition illuminates whatsoever it touches.

It seems trite to add that these remaining 16 chapters treat the existing knowledge of all that bears on physical limnology as nearly exhaustively as can be done usefully. To try to express the content of each chapter within a review one would have to condense Hutchinson's summaries. That cannot be done without loss of substance.

These chapters do, however, follow a general pattern though with a flexibility appropriate to the subject matter and the kind and quantity of existing information. Wherever possible he discusses the theoretical aspects first and then the empirical. He evidently agrees with the astronomer who said "there is little point in discussing facts until you have a theory". Doubtless understanding comes quicker if one has some sort of fabric to hold observations together. This is a habit of thought more geologists could cultivate with profit. Moreover, such a habit has the advantage of forcing one to quantify.

One other general pattern runs through the treatise and that is the practice

of pulling together scattered data, particularly data from sources not commonly available, converting them to the same units, and presenting them in tables or graphs. Two examples are Table 36 (p. 371), Estimates of total direct radiation on the 15th day of each month at various latitudes, and Table 53 (pp. 496-500), Heat budgets of the adequately known lakes of the world. This latter title understates the content of this table, which includes not only the heat budgets but the summer and winter heat incomes, the latitude, altitude, area, and mean and average depth of each lake. Users will be grateful for these conveniences.

Several chapters prompt comment for special reasons. The chapter on the hydromechanics of lakes is undoubtedly the toughest in the book. Even the author suggests the non-mathematically minded may choose to skip certain parts. But to skip wading through the treatment of the hydromechanics is to run the risk of not learning about the explanation for wind slicks, the counter-rotating helices of water driven by the wind, and above all, to risk missing the account of the astonishing antimerism of the Portuguese-man-of-war—wind propelled animals that are asymmetric so as to stay in the upwelling between the water helices where plankton are most abundant and furthermore having the opposite asymmetry in the southern hemisphere because of the geostrophic effect on the wind driven helices. These are delightful patches of sun-drenched meadow in a landscape strewn with mathematical boulders.

The chapter on the properties of water is a gem that should be required reading for all geologists as well as biologists. Here Hutchinson has brought together all the scattered knowledge about the structure and properties of water and presented them with elegant clarity. Fascinating is the story of this remarkable liquid—the structural similarity of ice and tridymite, the quasi-crystalline and ever changing structure of liquid water, the why of its anomalous melting and boiling points, its density regime, and the range of its solvent powers.

The chapters on the inorganic ions of rain, lakes, and rivers and the silica cycle in lake waters should also be required reading for geologists.

One of the most important discoveries ever made in limnology came from an attempt to increase the productivity of a small lake by adding phosphorous in relatively large quantities. The phosphorous added, even though in large amounts, had no permanent effect on the biochemistry of the lake. The seston and the sediments soon locked up the excess phosphorous so that the total amount left for biological use and recycling was just what it had been before enrichment. Hutchinson says (p. 746) the lake "acts as a self-regulatory system returning to a steady-state condition determined by all the variables that operate within it, after one of these variables has been disturbed".

Tucked away in the treatise, in appropriate places, are numerous surprising facts. A few of these will suffice to give the flavor: That owing to the length of day, more summer radiation is delivered at the North Pole than anywhere else on earth (pp. 371-372). That the green color of lakes is due to light scattered upward from particulate matter including organisms but is not due to the chlorophyll in organisms. (p. 417). That there are horizontal rainbows

(p. 419). That rain has a pH of 4 to 5 (p. 549). That at depths of 4.5 m lake water can be 381 percent saturated with oxygen (p. 620). That *Anabaena cylindrica* excreted, in culture, a remarkable polypeptide that forms complexes with Fe, Cu, and PO_4—this not only is likely to have great ecologic significance, as he points out, but also may help explain why certain planktonic organisms concentrate various metals by factors of 1,000 or more.

The treatise contains faults, but as Eric Partridge said in commenting on the faults in Fowler's Modern English Usage (N. Y. Times Book Review, March 9, 1958) "of course it does! Every worthwhile book contains many faults, and every worthwhile writer commits them. Look how shockingly incorrect is much of Shakespeare's English!" Nevertheless, I found only one sentence (p. 899) whose meaning was fouled up by a tricky double negative. Typographical errors are few and most of them are in expectable places, i.e. in running, simple, non-critical parts of the text—places most easily overlooked in reading proof. A few graphs have titles so brief that they are not satisfying.

Some non-mathematical readers may be complimented by Hutchinson's use of "obviously" in some of the mathematical argumentation. But if they feel complimented by this assumption about their facility with mathematics, they may feel transcendentally honored when they come to p. 349 where he alludes to a long and, to me, awesome term as "of course the well known error function".

Of the treatise as a whole, I agree with a good friend who wrote me just after the book came off the press: "Have you seen Hutch's book, it's terrific".

W. H. BRADLEY

The Leibniz-Clarke Correspondence; edited and annotated by H. G. Alexander. P. lvi, 200. New York, 1958 (Philosophical Library, Inc. $4.75). —The correspondence between Leibniz and Clarke is one of the most interesting documents in the philosophy of science. It is the controversy of two scientific giants (Leibniz and Newton) of the 18th century concerning natural religion, the nature of space, God's relation to the universe, the principle of sufficient reason, the meaning of force and other philosophical problems.

Alexander's introduction serves admirably as a summary of and a guide through the series of letters; it also provides the links with modern scientific issues which make the book a very timely one.

In November 1715, Leibniz warned his friend, the Princess of Wales, of the threat to natural religion that was inherent in Newton's philosophy. The princess attempted to initiate a literal exchange of views between the two great minds. However Newton showed himself unapprochable and selected as deputy and spokesman Samuel Clarke, a man greatly distinguished as a scientist versed in Newton's physics, and a theologian. Thus began what Voltaire called "peut-être le plus beau monument que nous avons des combats littéraires."

HENRY MARGENAU

Publications Recently Received

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